

Imperial College London

Department of Epidemiology and Biostatistics

Micro-environmental Models of Human Exposure to Air Pollution

PhD Thesis

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the degree of Doctor of Philosophy at Imperial College London*

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Declaration of Originality

I, Gioia Mosler, hereby declare that the work presented in this thesis, 'Micro-environmental Models of Human Exposure to Air Pollution', is my own. Where material and information is sources from outside this thesis it is clearly indicated and referenced.

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This work is dedicated to my family.

Abstract

Particulate air pollution (PM) has been shown by many studies to cause adverse health effects. Traditionally PM exposure was estimated using ambient concentrations. Lately, studies have revealed that this approach poorly reflects differences between individual's exposures and as such results in exposure misclassification.

This thesis aims to improve personal exposure predictions by building a model (MEPEX model), which takes into account the temporal and spatial variability of ambient PM, as well as visited microenvironments. For the composition of this model, existing approaches for model components were evaluated, compared and developed. A temporally adjusted land-use regression (LUR-adj) model for predictions of ambient $PM_{2.5}$ and PM_{10} was built, validated, and compared to estimates from a dispersion model. Ratios were developed to adjust ambient concentrations for cycling and in-bus transport microenvironments. Additionally, modelling approaches for the home indoor microenvironment were compared, using monitoring data. A secondary aim was to evaluate the performance of different approaches for personal exposure assessment by comparing varying levels of model sophistication.

Validation of the LUR-adj model showed good model fit ($IA > 0.5$) and low error ($NMSE < 1$) for short-term predictions of $PM_{2.5}$ and PM_{10} at locations in London. In comparison to predictions of a dispersion model (ADMS-urban), LUR-adj estimates of PM_{10} produced better results for model performance parameters at the majority of 26 predicted locations.

MEPEX model predictions of monitored daily personal exposure for an individual in London resulted in an R^2 of 0.439 for $PM_{2.5}$ and 0.403 for PM_{10} . Predictions using modelled home outdoor concentrations in comparison were lower with R^2 of 0.173 for $PM_{2.5}$ and 0.086 for PM_{10} . These results provide the first quantifiable evidence that personal exposure models of $PM_{2.5}$ and PM_{10} can reduce exposure misclassification compared to estimates based only on ambient PM.

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1. Introduction

1.1. Rationale

Air pollution exposure to particulate matter (PM) has been linked to several severe health effects, such as increased number of myocardial infarction and lung cancer. Studying people's exposure to PM is therefore important. Many connections between PM exposure and specific health effects were established in the 1980s and 1990s by applying simple estimates for pollution exposure, such as measurements at a central monitoring site in a study area. Temporal changes of air pollution concentrations could then be compared to temporal changes in health outcomes for a population in the area (Dockery et al. 1993; Brunekreef & Holgate 2002).

Depending on study design, individual differences between exposures in a population can be important, for example between members of a study cohort. Especially for large study populations, exposure of individuals is often approximated using monitored concentrations at a nearby site or by using models to predict outdoor (ambient) concentrations at the home address. Measurements at a nearby monitoring site, have however been shown to misrepresent exposure for a large part of the population (Avery et al. 2010; Özkaynak et al. 2013; Wilson & Brauer 2006). Even concentrations directly measured at home outdoor locations have shown little agreement with actual exposure of individuals, experienced during their personal daily routine, also called personal exposure. A meta-analysis by Avery et al. (2010) compares PM_{2.5} concentrations between monitored home outdoor and personal exposure from nine studies, representing 329 non-smoking participants. The median of the average Pearson's correlation for these studies between home outdoor concentrations and personal exposure concentrations is 0.54, ranging between 0.09 and 0.83. These results show that only a part of personal exposure can be explained by concentrations measured at home outdoor locations. Much of the variability of personal exposure remains unexplained by ambient concentrations.

Differences between personal exposures, which are not related to ambient concentrations, have been attributed to different locations and situations visited during the day. One study by Dons et al. (2011) compared monitored exposure of couples for which one partner of each couple is a home-maker and the other partner spends several hours a day at work. The collected concentrations of black carbon varied between the partners by up to 30%, despite the fact that they lived in the same home. This shows that differences in activities during the day can have a substantial impact on personal exposure. During the day most people pass through a range of different places and

situations for which PM concentrations differ compared to concentrations outdoors at home. Some types of commonly visited places create a specific pollution environment influenced by distinctive, to some extent homogeneous pollution concentrations, these are called microenvironments (Borrego et al. 2006). Commonly distinguished microenvironments include indoor microenvironments, such as home or work, and different transport microenvironments, such as cars or buses (Klepeis 1999; Özkaynak et al. 2008). Exposure to PM varies widely between microenvironments and specific situations within microenvironments, such as cooking, with sometimes high peak concentrations. These concentration differences are in fact often greater than temporal differences observed for ambient concentrations (Klepeis 1999; Lai et al. 2004; Long et al. 2000).

Several studies have developed approaches to predict personal exposure by combining exposures experienced at various times in different microenvironments. The approach usually combines a base-model for ambient concentrations with adjustments using models for commonly visited microenvironments. Estimates for different microenvironments are then time-weighted according to people's time-activities throughout the day. These so called personal exposure models have the potential to provide a much more realistic individual-level exposure estimate compared to proxies for personal exposure based on home outdoor locations. Several studies during the past few years have shown the potential of these types of models in exposure assessment. Borrego et al. (2006) for example developed a personal exposure model for the population of Lisbon, Portugal. Another model was developed by Dons, Van Poppel, Kochan, et al. (2014), calculating personal exposure for the population of Flanders, Belgium.

So far however, approaches to the development of a personal exposure model vary greatly. Differences include, for example the type of model chosen for predictions of ambient concentrations, the number and type of included microenvironments, the models or adjustments chosen to predict exposure in each microenvironment, as well as differences in temporal and spatial accuracy. Little is known about how these different approaches compare or about the importance of each of the included model components. Also, few of these models have been applied to and compared with monitored personal exposure for validation. In addition, only some of the existing models provide a transferable modelling approach, which could be applied in a different context.

Future development of these approaches would clearly benefit from an exhaustive review, comparison, validation and development of each of the model components. Furthermore, few of the existing personal exposure models include both predictions for ambient concentrations with high temporal and spatial resolution, as well as a high level of detail for microenvironmental adjustments. Such a detailed approach could more accurately reflect differences between personal exposures, yet existing models have not been validated with monitored personal exposure concentrations. In

addition, a comparison between the different methods used for the prediction of personal exposure could provide important information on the strengths and weaknesses of each approach.

1.1. Aims and Objectives

The aim of this thesis was to build an integrated model for personal exposure assessment taking into account the temporal and spatial variability of ambient particulate matter, as well as microenvironments. A further aim was to evaluate the performance of different approaches for personal exposure assessment, comparing varying levels of model sophistication. In order to achieve the aims of this thesis, the following specific objectives have been defined:

- Review the state of the art on exposure assessment and exposure models;
- Undertake bespoke ambient, personal, and microenvironmental air pollution monitoring campaigns to both explore levels of variation in exposures and provide data to calibrate and validate models that are developed and adapted for this study;
- Apply an existing land-use regression model for London and the Thames Valley region, to predict ambient concentrations using a GIS.
- Develop temporal adjustments for the long-term ambient concentration predictions by comparison and selection of different adjustment methods using air pollution monitoring data;
- Evaluate performance of the temporally adjusted land-use regression model for monitored locations in London and compare predictions to results of a dispersion model;
- Calculate and evaluate adjustments for in-transport microenvironments using monitoring concentrations collected in a bespoke transport monitoring campaign;
- Apply, validate and compare several different types of models for indoor microenvironments using monitored concentrations collected in an indoor-outdoor monitoring campaign;
- Integrate selected microenvironmental models with the modelled ambient concentrations to predict personal exposure;
- Validate the modelling approach using data from the personal exposure monitoring campaign;
- Compare predictions from the personal exposure model with estimates from modified model variations;
- Demonstrate application of the model to predictions of exposure for a population in London.

1.2. Thesis Structure

Chapter 2 gives an introduction to particulate air pollution, its health effects and exposure.

Furthermore, different approaches to exposure modelling from the literature are explained with a focus on personal exposure models. Components of personal exposure models are then reviewed in detail, as well as potential approaches to integrate these components into a personal exposure model.

Chapter 3 introduces different the air pollution monitoring campaigns undertaken as part of this thesis work. Data collected during these campaigns is then described and. Finally, the preparation of the monitored data for the following analyses is explained.

Chapter 4 describes the development of a temporally adjusted land-use regression model to predict ambient concentrations with high temporal and spatial accuracy. First, the development of a land-use regression model as part of the ESCAPE study is described. Then, different approaches from the literature to temporally adjust the long-term predictions are introduced, compared, and selected. Finally, the temporally adjusted land-use regression model is validated for short-term predictions of locations in London and results are compared to predictions from a dispersion model.

Chapter 5 outlines the preparation of a set of microenvironmental models, adjustments, and estimates for the integration into a personal exposure model. Ratios and estimates to adjust for transport microenvironments are calculated from a transport monitoring campaign undertaken as part of this thesis, or alternatively selected from the literature. For adjustments of ambient concentrations to predict indoor residential exposure, different existing models from the literature are selected. The models are applied and compared for the prediction of monitored indoor concentrations, using data collected as part of this thesis for indoor and outdoor residential concentrations. Furthermore, an adjustment for in-office exposure is selected from the literature.

Chapter 6 describes the integration of the model of ambient concentrations, as well as the microenvironmental models or adjustments with individual time-activity to develop a personal exposure model. Model performance is then evaluated for predictions of monitored personal exposure collected for this thesis. The performance is furthermore compared to predictions of other approaches for to personal exposure assessment for the monitored personal exposure of an individual in London. Finally, model application is demonstrated for a population in London.

Chapter 7 summarises results from this thesis and reflects about on the implications of the findings in the wider context of air pollution exposure assessment.

2. Background and State of the Art

2.1. Introduction to Particle Exposure

2.1.1. Particulate Matter

Several different types of air pollutants have been connected with adverse health effects, including for example particulate matter (PM), ozone (O₃), nitrogen oxides (NO_x), sulphur dioxide (SO₂), volatile organic compounds (VOCs) (World Health Organisation 2006). This PhD focuses on PM, which is associated with some of the most severe air pollution related health risks.

PM comprises of airborne particles (also referred to as 'particles'), which originate from a variety of substances and/or sources. PM can be in either a solid or liquid state and vary in its composition. The composition of PM depends on many different factors, including source, climate, and the topography of the location. PM originates either from natural sources or from sources related to human activity (Gehring et al. 2002). Naturally occurring PM includes for example dust from the earth's surface, sea salt, and biological material. Particles from human activity come to a great extent from combustion processes, for example of cars, factories and domestic fires, but also from non-combustion sources, such as tyre and brake wear on vehicles or re-suspension of road dust. Some of these components are more toxic than others, a higher toxicity has for example been shown for particles from combustion (World Health Organisation Europe 2004; Brunekreef & Forsberg 2005; Perez et al. 2009). Despite the different composition and toxicity, sample size fractions are considered to have many properties in common. Size for example determines physical properties, such as deposition. Particles of the same size also have the same ability to penetrate into the respiratory tract or lung (Health Effects Institute 2002). Size fractions are therefore most commonly used to categorise PM. Size fractions are defined by the aerodynamic diameter of the particles in micrometres (µm) and are usually measured as mass in µg/m³. PM sizes generally vary between 0.005µm and 100µm in aerodynamic diameter (Health Effects Institute 2002). Table 1 shows some of the most commonly used particle size fractions: PM_{0.1}, PM₁, PM_{2.5}, PM_{10-2.5}, PM₁₀, and total suspended particles

Size	Name
$\leq 0.1\mu\text{m}$	PM _{0.1} or 'ultrafine particles'
$\leq 1\mu\text{m}$	PM ₁
$\leq 2.5\mu\text{m}$	PM _{2.5} or 'fine particles'
Between $2.5\mu\text{m}$ and $10\mu\text{m}$	PM _{2.5-10} or 'coarse particles'
$\leq 10\mu\text{m}$	PM ₁₀
All airborne particles	'Total suspended particles' or TSP

Table 1: Commonly used size fractions of airborne particles.

Measurement of black or British smoke (BS) and the coefficient of haze (COH) involve relatively simple but not very precise methods, based on degree of blackening of air pollution measurement filters and do not distinguish between different sizes of PM. The term respirable particles has been used by some studies, but does not refer to a consistent size fraction (Gee & Raper 1999; Nyberg et al. 2000). Most studies today therefore work with size fractions, as presented in Table 1. This thesis focuses on PM_{2.5} and PM₁₀, two of the most commonly studied particle size fractions (World Health Organisation 2006). Most of PM_{2.5} is created by combustion processes. Coarse particles (PM_{10-2.5}) on the other hand are often broken down from larger materials in mechanical processes. The distribution of fine and coarse particles also differs (Brunekreef & Forsberg 2005; Pope III & Dockery 2006). In addition, suspension of particles is also influenced by their size and mass. Heavy particles have a higher tendency to deposit, while lighter particles remain suspended for longer.

Much of PM is in the form of directly emitted particles, which are also called primary particles. Quite a significant part of PM however is secondary in origin. Secondary particles are produced by chemical reactions in the atmosphere. Nitrogen oxides, sulphur dioxide and organic compounds react, for example, with ozone to form new particles such as nitrates or sulphates (Bell et al. 2004; Malcolm et al. 2000).

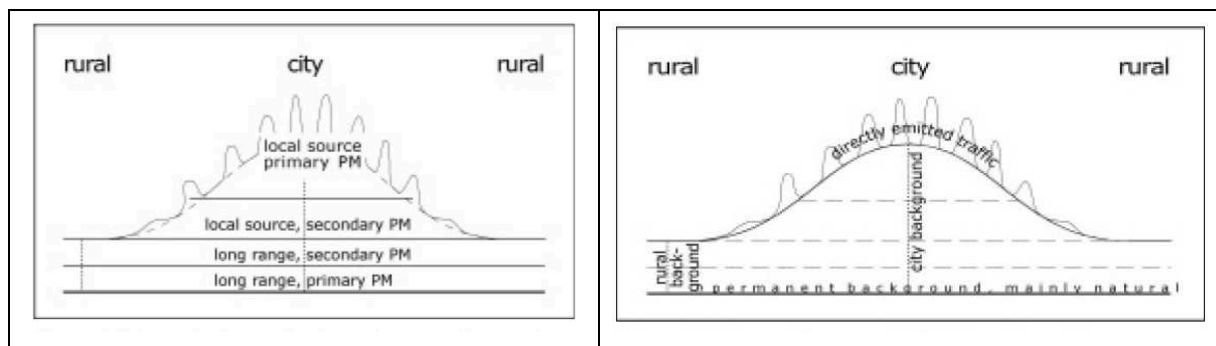


Figure 1: Schematic view of a city and its surrounding rural area, showing left: short range, long range, primary and secondary pollution, and right: emissions from traffic, city and rural background concentrations. Source: Mosler (2008)

PM levels vary substantially over space as well as over time. In terms of its distribution one can distinguish local or short range PM and long range or far travelled PM (Bell et al. 2004). Short range PM is derived by local sources, mainly combustion, and is therefore, for example high next to major roads, and levels drop off rapidly with increasing distance from the source. Long range PM on the other hand is much less influenced by proximity to sources, and can travel as part of an air mass over hundreds of kilometres (Buchanan et al. 2002; Malcolm et al. 2000). The difference between long range and short range, as well as primary and secondary particles can be observed most prominently in cities due to the large number of source activities (from vehicles, domestic heating etc.), see also Figure 1 (left). Malcolm et al. (2000) has for example shown that on average 75% of the sulphate component of particles measured at UK monitoring stations originates from the UK, whereas 25% originates from outside the UK. In locations in the southeast of England, including London about 60% derive from inside and 40% from outside the UK. Results by Lenschow et al. (2010) show that, for Berlin, about 55% of the urban background PM₁₀ pollution could be attributed to long range PM. Urban background, as referred to by Lenschow et al. (2010) comprises of all particles that can be observed in a city away from the direct influence of traffic sources (see Figure 1, right side).

2.1.2. Health Effects Associated with Particles

Human exposure to PM has been linked to a number of different adverse health effects, increased numbers of emergency room visits, hospital admissions, and increased mortality. Some of the main health effects related to particle pollution are listed in Table 1. The list is based on evidence summarised by the World Health Organisation Europe (2004). The table shows that severe effects have been observed both for short-term (e.g. daily) and long-term (e.g. annual) exposure to increased particle concentrations. Associations of a range of different respiratory health effects have been found, such as reduction in lung function. Additionally there is evidence for severe effects on the cardiovascular system, such as increases in myocardial infarction (World Health Organisation Europe 2004).

Recently the International Agency for Research on Cancer (IARC) has additionally classified PM as carcinogenic, Group 1 (International Agency for Cancer Research 2013), meaning that there is “sufficient evidence of carcinogenicity in humans”. The IARC is an international body which publishes an extensive series of monographs on carcinogenic risks to humans. Within these monographs, carcinogenic risks related to airborne particles have been mentioned for example as part of the carcinogenic potential of Polycyclic Aromatic Hydrocarbons (PAH) in volume 92 and as part of indoor combustion in volume 100E. A complete monograph on the carcinogenicity of

outdoor air pollution and PM is planned for future publication (<http://monographs.iarc.fr/ENG/Classification/ClassificationsAlphaOrder.pdf>, accessed: 4th Aug. 2014).

Short-term Effects	Long-term Effects
• Lung inflammatory reactions • Respiratory symptoms (including Asthma episodes²) • Adverse effects on the cardiovascular system • Increase in medication usage • Increase in hospital admissions • Increase in mortality • Lung inflammatory reactions	• Increase in lower respiratory symptoms • Reduction in lung function in children • Increase in chronic obstructive pulmonary disease • Reduction in lung function in adults • Reduction in life expectancy, owing mainly to cardiopulmonary mortality and lung cancer • Increase in lung cancer¹

Table 2: Effects of increased particle exposure. Main source: World Health Organisation Europe (2004), ¹ (International Agency for Cancer Research 2013), ² Delfino et al. (1998; 2002)

Evidence linking lung cancer in particular to increased PM concentrations has been established by Raaschou-Nielsen et al. (2013). The study compared long-term home outdoor PM concentrations for participants of 17 European cohorts with lung cancer incidents. The meta-analyses showed a statistically significant association between the risk for lung cancer and PM. The study was undertaken as part of ESCAPE (European Study of Cohorts for Air Pollution Effects), a 7th framework EU project. Several sections of this thesis have been conducted as part of the ESCAPE project by the PhD candidate.

The ESCAPE project included extensive monitoring campaigns of PM_{2.5} and PM₁₀, as well as development of air pollution models in 22 different European centres. The effect of particle exposure on health endpoints for a large number of European cohorts was then evaluated. In the following paragraphs some of these studies are introduced.

Respiratory health effects were assessed for example as part of a study on lung function in children by Gehring et al. (2013), who found a small decrease in lung function associated with increased PM_{2.5}, but not with PM₁₀. Varying results for the association between PM and lung function were furthermore addressed in a study by Eeftens et al. (2014), who examined the effect of different elemental composition on lung function, as a potential cause for the heterogeneity in the results of the study. Eeftens et al. (2014) could however not find a strong association with a particular element.

One ESCAPE study by Macintyre et al. (2014) looked at the effects of air pollution on respiratory infections during early childhood. The study found a statistically significant increase in pneumonia for increased levels of PM₁₀, but not for PM_{2.5}. A further study by Fuertes et al. (2014) showed that the effect of PM on pneumonia could not be explained by a particular PM element.

One further study on respiratory health was conducted as part of the ESCAPE study, for which the PhD candidate is co-author (Schikowski et al. 2014). The study focuses on COPD (chronic obstructive pulmonary disease) and showed a positive association between PM_{2.5} and PM₁₀ concentrations at home and COPD prevalence in the cohort populations. The results were however not statistically significant.

Several studies on cardiovascular health were conducted as part of the ESCAPE project. Arterial blood pressure was assessed in a study by Fuks et al. (2014), which elevated blood pressure was associated with increased concentration of particles. Only results for PM_{2.5} were statistically significant, no association was found for PM₁₀. A statistically significant association between PM and acute coronary events could be established in a study by Cesaroni et al. (2014). The findings were based on 11 cohorts and showed that a 5µg/m³ increase in PM_{2.5} was associated with a 13% increased risk of coronary events. For PM₁₀, a 10µg/m³ increase in estimated annual concentrations was associated with a 12% increased risk of coronary events.

A recent, contemporaneous field of research examines the link between PM exposure and cerebrovascular events or stroke. Studies on increased risk of ischaemic stroke in relation with increased short-term particle concentrations had shown contradictory evidence (Wellenius et al. 2012; O'Donnell et al. 2011). Results from the ESCAPE study based on long-term particle exposure of 11 cohorts have associated a 5µg/m³ increase in annual PM_{2.5} exposure with 19% increased risk of incident stroke. Similar findings were obtained for PM₁₀ (Stafoggia et al. 2014). In another study conducted as part of the ESCAPE project increased PM concentrations were additionally associated with cerebrovascular mortality (Beelen, Stafoggia, et al. 2014)

Recently, research conducted as part of the ESCAPE project, as well as by other studies has shown that exposure to PM during pregnancies is associated with restricted foetal growth and low birth weight (Pedersen et al. 2013; Slama et al. 2007). Another study conducted as part of the ESCAPE project additionally associated PM exposure during pregnancies with delayed psychomotor development during childhood (Guxens et al., 2014).

Historically, one of the first health effects associated with increased PM exposure was mortality, for example as part of the 'Harvard six cities study' (Dockery et al. 1993). Associations between PM

exposure and natural-cause mortality was confirmed by results from the ESCAPE study (Beelen, Raaschou-Nielsen, et al. 2014) based on 22 cohorts.

Results of studies conducted as part of the ESCAPE project show that the association between PM and health effects can differ between studies. The summary of health effects by the WHO (see Table 2) for example includes reduced lung function in children for increased PM_{2.5} and PM₁₀ exposure. Results from the ESCAPE project by Gehring et al. (2013) on the other hand only show a significant association for PM_{2.5}, not for PM₁₀. This result also highlights another issue, namely that health effects associated with PM_{2.5} and PM₁₀ may differ. In addition, heterogeneous results between different cohorts have been observed for the relationship between PM exposure and COPD prevalence by Schikowski et al. (2014).

Differences between results can occur for a range of reasons, but mostly relate to two main areas of potential error. The first area is related to non-PM related influences on the prevalence/incidence of the studied health endpoint. These influences can for example include socioeconomic differences, age differences, or the degree of exposure to other relevant pollutants. Studies of the ESCAPE project accounted for a range of these potential confounders in their analyses. Some further confounders, for example related to cultural or lifestyle differences are more difficult to quantify and may result in residual confounding (Gruziova et al. 2014; Beelen, Raaschou-Nielsen, et al. 2014; Raaschou-Nielsen et al. 2013).

The second area is related to the PM exposure attributed to each individual. Exposure for the ESCAPE study and most other larger cohort studies is based on home outdoor PM concentrations. Most people however spend several hours away from home every day and are influenced by concentrations in different MEs. Home outdoor concentrations can therefore not fully reflect their personal exposure and as such result in exposure misclassification for a large part of the population (Özkaynak et al. 2013; Dons et al. 2011). Exposure misclassification is further examined later in this chapter.

Health effects have been studied by the ESCAPE project with a focus on long-term PM exposure. One main objective of this thesis is to develop a model which can predict individual short-term variations in exposure. Short-term health effects are therefore of particular interest to this study (see summary in Table 2, left). A number of studies have shown an association between daily changes in air pollution levels and adverse health effects to the cardiovascular and respiratory system (Peters et al. 2001; Maynard et al. 2007; Törnqvist et al. 2007; Wellenius et al. 2012; Pope III & Dockery 2006). Health effects of exposure to air pollution for even shorter time periods, such as hourly or for several hours have also been confirmed (Michaels & Kleinman 2000). A study by Peters et al. (2001) showed

elevated risk of myocardial infarction within a few hours of observed elevated PM_{2.5} concentrations. Increases in blood pressure in elderly people were observed at a five hour lag for increased PM_{2.5} concentrations by Harrabi et al. (2006). A study by Wellenius et al. (2012) found greatest increased risk of ischaemic stroke within 12 to 14 hours of increased PM_{2.5} exposure. Effects of coarse particles on heart rate have also been shown for sub-daily concentration increases (Alexis et al. 2006; Ljungman et al. 2008).

Many studies have focused on establishing health effects either for PM_{2.5} or for PM₁₀ and similar types of health effects have been associated with either size fraction (World Health Organisation Europe 2004; World Health Organisation 2006). Fine (PM_{2.5}) and coarse (PM_{10-2.5}) particles have however different origin and distributions. Some studies therefore investigated the question of whether fine (PM_{2.5}) and coarse particles (PM_{10-2.5}) show different effects on health. A review by Brunekreef & Forsberg (2005) found that several studies show effects for the coarse particle fraction that were different from PM_{2.5}. Toxic effects have however been shown for both the coarse and the fine particle fraction (Health Effects Institute 2002; Brunekreef & Forsberg 2005). The studies underlying these conclusions work on a mass basis, and other findings show that when particle number and surface area are taken into account, fine particles would have a higher toxicity than the coarse fraction (Health Effects Institute 2002). Evidence, such as that given by Gordon (2007) suggests that different PM sizes and sources are responsible for certain distinct cardio-respiratory health effects. For example, components of smaller size can penetrate deep into the lungs; particles with sizes above 10µm on the other hand deposit almost exclusively in the nose and in the throat.

For several health outcomes the effect of certain particle size fractions could be quantified. Dose-response rates show a relationship between a certain incremental step in air pollution concentration with a certain increase in health effect. Dose response rates can for example be applied to estimate health effects for certain scenarios, e.g. what would be the decrease or increase in adverse health effects if traffic regulations were changed in a city (World Health Organisation Europe 2006a). A study by Schwartz, Dockery, & Neas (1996) calculated from data collected for the Harvard six cities study showed that an increase of 10µg/m³ in PM_{2.5} was associated with a 1.5% increase in total daily mortality. A study by the World Health Organisation Europe (2013) reviewed dose-response relationships for PM₁₀ from the literature and derived a relative risk for mortality of 1.026 for each 10µg/m³ increase for an adult population.

Based on knowledge about the connection between a particular pollutant and health effects, limit concentrations were calculated, which are used to govern concentration levels for an area, as well as to issue public health warnings for exceedances. During the last decades many governmental bodies and NGOs (Non-Governmental Organisations), such as the WHO (World Health Organisation) have

established limit values for air quality standards of PM₁₀ and to some extent for PM_{2.5}. For PM₁₀ a daily average concentration of 50µg/m³ is given as limit value by the WHO (World Health Organisation 2006) above which concentrations are considered harmful to the public. The Department of Environment, Food, and Rural Affairs (DEFRA) established the same limit in the UK. In addition, DEFRA implemented air pollution bands (incremental steps of concentration), which are related to health warnings (see Table 2, as published by the London Air Quality Network – LAQN: <http://uk-air.defra.gov.uk/air-pollution/daqi?view=more-info&pollutant=pm25#pollutant>).

For PM_{2.5} there are less air quality guidelines in place. A daily average of 25µg/m³ is given as a limit value by the WHO (World Health Organisation 2006). The daily limit for PM_{2.5} in the US was set by the Environmental Protection Agency (EPA) at 35.4µg/m³ (Environmental Protection Agency 2012). The UK DEFRA similarly has applied 35 as the upper limit of the low pollution band, see Table 3 (<http://uk-air.defra.gov.uk/air-pollution/daqi?view=more-info&pollutant=pm25#pollutant>, accessed: 22.3.2014).

Banding	PM _{2.5} (µg/m ³)	PM ₁₀ (µg/m ³)	Accompanying health messages for at-risk groups and the general population	
			At-risk individuals*	General population
Low	<35	<50	Enjoy your usual outdoor activities.	Enjoy your usual outdoor activities.
Moderate	36-53	51-75	Adults and children with lung problems, and adults with heart problems, who experience symptoms, should consider reducing strenuous physical activity, particularly outdoors.	Enjoy your usual outdoor activities.
High	54-70	76-100	Adults and children with lung problems, and adults with heart problems, should reduce strenuous physical exertion, particularly outdoors, and particularly if they experience symptoms. People with asthma may find they need to use their reliever inhaler more often. Older people should also reduce physical exertion.	Anyone experiencing discomfort such as sore eyes, cough or sore throat should consider reducing activity, particularly outdoors.
Very High	>70	>100	Adults and children with lung problems, adults with heart problems, and older people, should avoid strenuous physical activity. People with asthma may find they need to use their reliever inhaler more often.	Reduce physical exertion, particularly outdoors, especially if you experience symptoms such as cough or sore throat.

Table 3: Air pollution bandings for PM_{2.5} and PM₁₀ with respective concentration limits and health warnings for individuals at risk, as well as for the general population. Source: LAQN (<http://www.londonair.org.uk/london/asp/airpollutionhealth.asp?HealthPage=HealthAdvice> and <http://www.londonair.org.uk/london/asp/airpollutionindex.asp>, both accessed: 16th April 2014) * Adults and children with heart or lung problems

The implemented limit values, as recommended by the WHO, the EU, DEFRA and other official bodies, clearly fulfil an important role when issuing public health warnings or when used as a regulatory tool. The application of limit values however has several clear limitations:

- 1) Health effects have been shown to be continuous and do not stop when a certain threshold is reached (Wellenius et al. 2012; World Health Organisation Europe 2006b; Bayer-Oglesby et al. 2004).
- 2) Applied limit values are calculated for outdoor air pollution concentrations. Concentrations of personal exposure however have shown to be different from concentrations outdoors (Avery et al. 2010). Limit values for actual personal exposure concentrations may therefore also differ.
- 3) Applied limit values are intended as a political tool and to some extent related to political context. Daily limit concentrations for PM₁₀ in China for example are three times higher than in the EU (O'Keefe 2013).

Epidemiological Studies

Many epidemiological studies have been conducted in order to understand the relationships between particulate matter and health effects (Brunekreef & Holgate 2002; World Health Organisation Europe 2004; Pope III & Dockery 2006). Traditionally those health assessment studies focus either on short-term (e.g. hours, days or weeks) or long-term (e.g. years) exposure.

Short-term associations between pollution levels and acute health outcomes are traditionally assessed with time-series studies (Amman et al. 2005; Bell et al. 2004). Time-series studies refer to study designs which relate temporal changes in pollution concentrations to temporal changes in health endpoints of the studied population. The Harvard six cities study provided crucial evidence in establishing the relationship between increased particle exposure and mortality (Dockery et al. 1993) by comparing PM₁₀ concentrations measured at central sites in six cities in the USA with daily mortality.

The relationship of long-term health in a given population or population group to air pollution is usually assessed by looking at geographical differences in cohort studies (World Health Organisation Europe 2004). Cohort studies in this context refer to studies which compare differences in pollution concentrations at an individual's homes to differences in health between individuals of a population cohort.

Both types of studies have proven to be useful tools to assess the health risk of a population. Neither method can however include the full range of particle variability over space and time. By their very design time-series studies neglect geographical variation and cohort studies neglect variation over

time. Additionally, differences in time-activity-patterns within a population, or exposures in specific microenvironments (i.e. at home inside) are usually not considered (Bayer-Oglesby et al. 2004). The exposure of individuals may differ considerably from long-term averages outside at their home location. In addition, temporal variability of some people's exposure during their daily routine may be poorly reflected in the short-term variability at a nearby monitoring site. More refined models for personal exposure have therefore been developed. These models include exposure differences for microenvironments passed by individuals, and in addition may, depending on the study design, work with high temporal as well as spatial accuracy.

2.1.3. Exposure to Particles

Three main terms are used to describe levels of air pollution: concentration, exposure, and dose (or intake). 'Concentration' is the level of a pollutant at a certain place and time. Human 'exposure' to a pollutant is defined as: "... the event when a person comes into contact with a pollutant of a certain concentration during a certain period of time" (Bayer-Oglesby et al. 2004). The 'dose' or intake of a pollutant refers to the amount of pollution that entered the body by inhalation (Jensen 1999). The amount of an air pollution dose of an individual may as such be different from the exposure, depending on rate of inhalation.

Variability of particle concentrations and exposure

Air pollution concentrations vary in space and time, not only outside, but also within different MEs (such as home indoors or inside a car). How particles vary over space depend on the following factors. Concentrations are high directly next to sources and disperse quickly (within meters) away from sources. The immediate dispersion of particles depends largely on wind speed and direction, as well as potential obstacles for air flow. Principles for inner- and inter- regional air pollution distribution furthermore depend on larger climatological influences, for example temperature, air pressure, as well as direction and speed of large air masses. These influences are on the ground modified by properties of the earth's surface, for example air travels faster over smooth surfaces, such as water bodies, compared to rough surfaces, such as cities (Cambridge Environmental Research Consultants Ltd. 2013). In addition, particle concentration decreases with height (Kaur et al. 2007). Temporal variability of particle concentrations has been described above. Temporal changes generally depend on locally varying source activity, as well as concentration changes due to long-range particle transport. Spatial and temporal changes in concentrations have so far only referred to ambient pollution. Most people however spend a less time outdoors and a lot of their

day in specific microenvironments. Microenvironments (MEs) are places where local conditions create a homogeneous pollution compartment distinct from its surrounding. They are determined by the types of sources inside the compartment and by factors determining the influence from sources outside (Borrego et al. 2006; Gerharz et al. 2013; Hertel et al. 2001). Concentration changes in MEs are in most cases influenced by ambient air, but influences in addition include source activities and dynamics within the ME. A commonly studied ME is for example home indoors (Hoek, Kos, et al. 2008; Hänninen et al. 2004). Home indoor MEs show, to some extent concentrations which penetrated from the outdoors, but are in addition influenced by indoor sources, such as cooking or cleaning. A more detailed description of air pollutant concentrations in different MEs is given below.

Quantifying exposure using the direct and indirect method

Two main methods have been used to quantify exposure: the direct (monitoring) and the indirect (modelling) method. The direct approach includes all studies that directly monitor the exposure of each individual in a study population. While this results in exact measurements of exposure to a pollutant (limited only by the quality of the measurements), it is practically impossible to monitor everybody. With an indirect approach, levels of pollution are indirectly estimated or calculated through models, monitored air pollution is however used for model development and model validation (Borrego et al. 2009; Moschandreas & Saksena 2002; Steinle et al. 2013).

Exposure misclassification:

Estimates of exposure are approximations of the actual exposure of individuals. The extent to which the exposure estimate and the actual exposure differ is called exposure misclassification (Bayer-Oglesby et al. 2004). The term 'exposure misclassification' has been used in particular in relation to traditional methods to estimate exposure variability in a population, either by only using temporal differences (e. g. at a central site) or by only using spatial variability (long-term averages at home addresses or nearby monitors). Focusing on temporal variability includes limited representation of spatial variability, focusing on spatial variability means limited representation of temporal variability for the study population. In addition, neither method accounts for variability related to different MEs visited by individuals during the day, or for differences between time-activity patterns. Therefore both methods result in exposure misclassification for a significant part of the population (Avery et al. 2010; Özkaynak et al. 2013; Sarnat et al. 2005). The importance of exposure misclassification however depends on spatial and temporal aspects, as well as the aims of each study. In addition, the degree to which refined exposure estimates (e.g. including location of

individuals) influence predictions of health outcomes depends on study-specific characteristics, such as studied health outcome, or particle size fraction. In time-series studies for example only population-level exposure estimates are needed, as the focus is entirely on quantifying a temporal effect. In individual level studies, such as cohort studies the variability between the individuals is more important (Özkaynak et al. 2013).

For studies that focus on sub-daily health outcomes (e.g. looking at onsets of cardiac arrests or stroke), reducing exposure misclassification is of particular importance, as exposure variability on an intra-day level depends to a large extent on exposure in different MEs (see ME descriptions below). In addition, different exposure situations and MEs may provide potential for exposure reductions, e.g. by change of commuter mode or route. For studies focused on individual exposure reduction, refined personal exposure models are therefore essential.

In the following sections first exposure monitoring, then exposure modelling are introduced in more detail.

2.1.4. Monitoring Particle Concentrations

Air pollution is usually monitored either at permanently installed sites or for bespoke monitoring campaigns for which temporally installed stationary sites or mobile devices are used.

Permanently installed (or fixed-site) monitors have played an important role in establishing links between concentrations at central monitors and health effects in the population living in the area. As mentioned before, data from fixed site monitoring stations are in addition utilised as a political tool to govern pollution levels, as well as to issue health warnings. A network of stationary PM monitoring sites is operated by DEFRA in the UK as part of the Automatic Urban and Rural Network (AURN). Cities or local authorities sometimes additionally run their own monitoring networks, in London this network is managed by the London Air Quality Network (LAQN) based at Kings College London with currently (May 2014) 27 stations monitoring PM_{2.5} and 74 stations monitoring PM₁₀.

Apart from networks using fixed monitoring sites, monitored air pollutant concentrations have also been collected by purpose-designed monitoring campaigns set up for specific studies. Three distinctive monitoring campaigns can be defined:

1. Outdoor monitoring at locations not covered by existing networks (de Hoogh et al. 2013),
2. Different microenvironments, such as home indoors or in a car,
3. Using personal monitoring devices carried by individuals for a period of time during certain activities.

Many studies have collected monitoring concentrations for indoor MEs, which has an important contribution to exposure, as we spend on average most of our time at home indoors (Schneider et al. 2004). Indoor monitoring is often done in conjunction with outdoor monitoring at the facade to study particle propagation from outdoor to indoor air (Meng et al. 2005). A more detailed description of the home indoor MEs is given below.

Monitoring in transport MEs is a further focus of ME monitoring (Kaur et al. 2006; Adams et al. 2001). Different transport MEs are generally defined for different transport modes (walking, cycling, in-car, in-bus, in-train, etc.). The dynamics of PM concentrations in transport MEs are particularly complex and only little is known about exposures in transport MEs. Most monitoring campaigns that have been undertaken in transport MEs are therefore highly specific and results can seldom be transferred to another person, time or area (McNabola et al. 2009b; Kaur et al. 2005a). A more detailed description of transport MEs is given below.

Personal monitoring is achieved by recording air pollution during part of a person's activities, while the monitor is kept close to the person. Personal monitoring is attempting to record the actual exposure of individuals. People's activities often take place in a certain set of MEs (such as indoor at home, in traffic, and at work) and are often collected for model evaluation (M. Toivola et al. 2004; Nerriere et al. 2005; Williams et al. 2008).

Air Pollution Monitors

A wide variety of monitoring systems are available to measure particle concentrations. For this study only devices that measure $PM_{2.5}$ and PM_{10} are of interest. The main criteria, which distinguish monitors for different practical applications are portability, temporal resolution, reliability and accuracy. Constraints in resources and availability inevitably also often play a major role when choosing a type of monitor for a study. The smallest devices are hand-held and can be carried around, whereas larger monitors are usually installed permanently (Figure 2). Some monitors provide PM estimates over a time period, such as 24 hours or longer. These devices often rely on filter based technologies in which particles accumulate on a filter. This method is also referred to as passive sampling or gravimetric sampling. One of the most commonly used passive samplers is the Harvard Impactor (de Hoogh et al. 2013; Zuurbier et al. 2010), which is described in further detail in chapter 3. The filters of passive samplers have to be regularly changed and are then analysed for number of particles or particle mass in a laboratory, further analyses may include identification of different chemicals. Many personal exposure studies are conducted using passive samplers, as they are often easier to handle. The long averaging time (days or weeks) makes investigations of MEs or

ME situations, such as indoor cooking, difficult (Gauvin et al. 2002; Hänninen et al. 2004; Harrison et al. 2002; Janssen et al. 1998; Janssen et al. 2005; Koistinen et al. 2001; Kousa, Oglesby, et al. 2002; Lai et al. 2004; Violante et al. 2006; Johannesson et al. 2011; Williams et al. 2008; M Toivola et al. 2004; Adgate et al. 2007; Monn et al. 1997).



Figure 2: Left side: A handheld Dustmate monitor from Turnkey (<http://www.turnkey-instruments.com>, accessed: 11th Sept. 2013); right side: a permanently installed monitor at a monitoring site in London. Source: London Air Quality Network (<http://www.londonair.org.uk>, accessed: 11th Sept. 2013).

Continuous monitors on the other hand provide short-term results, for example on a timescale of minutes. Most types of continuous monitors work with light or laser scattering, counting the particles that pass through a light or laser beam. These technologies are sometimes also referred to as optical methods. Examples of monitors using optical methods are Osiris and Dustmate Monitors (Turnkey Instruments Ltd., Northwich, UK), or DataRAM nephelometers (MIE Inc., Bedford, MA, USA). A further description of light-scattering methods with specific focus on Osiris and Dustmate monitors is given in chapter 3. A different technique for short-term particle measurements is used by aethalometers. These devices collect particles in a filter material, which creates deposits of increasing density. The opaqueness of the deposits is then measured by a light beam in short time intervals. Aethalometers are available as stationary instruments, as well as in portable sizes (<http://www.environmental-expert.com/products>, accessed: 21st May 2014).

Continuous monitors are frequently used for transport ME monitoring, as the high frequency of concentration changes in traffic requires high frequency monitoring. Lately continuous monitors

have increasingly been used as well for personal monitoring campaigns (Dons, Van Poppel, Kochan, et al. 2014).

Due to the differences in technology, concentrations measured by different devices are often not the same. Although several studies have tried to explain or at least quantify these differences, knowledge so far remains limited.

2.2. Modelling Personal Exposure

2.2.1. Introduction

Due to the limitations of individual monitoring of exposure, exposure modelling is crucial to understand and predict exposure to particle pollution (Hertel et al. 2001). A range of models have been developed to predict ambient particle concentrations (as explained in detail below). These models have been developed to predict concentrations of ambient particles for areas not covered by monitoring station networks and to produce spatially and in some cases temporally more refined predictions for areas compared to stationary monitors. Estimates for concentrations at the residential address can provide better predictions of population exposure compared to using monitored concentrations from nearby stationary monitors. Personal exposure has however been shown to be different from ambient concentrations outside the home (Avery et al. 2010). Personal exposure models have therefore been developed that do not only base exposure on predictions of ambient particle concentrations, but also include non-ambient components in the form of ME visited during periods of time (Dockery & Spengler 1981; Freijer et al. 1998; Gariazzo et al. 2014; Montagne et al. 2013; Dons, Van Poppel, Kochan, et al. 2014; Borrego et al. 2006; Kruize et al. 2003; Hänninen et al. 2003; Baklanov et al. 2007; Hänninen, Karppinen, et al. 2005; Kousa, Kukkonen, et al. 2002).

2.2.2. Predicting Personal Exposure with the Microenvironmental Approach

Personal exposure models estimate the exposure of an individual from different MEs, an approach also referred to as the microenvironmental approach (Chan et al. 1999; Jensen 1999). These models summarise exposure using the sum of visited MEs during a day, multiplied by the time spent in each one of them. The times in each ME are then time-weighted to get exposure estimates for example for different hours of a day. This exposure model (E) has been written as follows by Kruize et al. (2003):

Equation 1: Microenvironmental Approach

$$E = \sum_i^N f_i * C_i$$

Where C_i is the concentration in microenvironment i , f_i the fractional time spent in microenvironment i , and N the number of MEs. This microenvironmental approach has been applied in many studies and has also been referred to as ‘integrated modelling approach’ by some studies (Baklanov et al. 2007; Steinle et al. 2011; Briggs et al. 2005), as it combines ambient concentrations with time-activity data and different ME models.

In the following paragraphs several studies are reviewed, which developed and in some cases applied integrated personal exposure models for particle pollution. A focus on European studies is taken, as lifestyle, built environment, building design, population density, and city layout are relatively similar within Europe, but different from other parts of the world. For example common use of air conditioning for buildings and vehicles is especially important in Northern America (Gupta & Cheong 2007; Thornburg et al. 2001; Croxford et al. 2000). Use of air conditioning however substantially alters indoor particle concentrations (Hänninen 2005; Ho et al. 2004; Gupta & Cheong 2007; Horemans et al. 2008; Ní Riain et al. 2003). One US-American model is however of major importance in terms of its general approach: The SHEDS (Stochastic Human Exposure and Dose Simulation) model, developed by the US EPA is a temporally and spatially transferable modelling framework that has been applied by several studies. It provides parameters for indoor modelling and estimates for other MEs, which can be combined with modelled or monitored ambient concentration studies (Burke et al. 2001; Özkaynak et al. 2013; Georgopoulos et al. 2005). No equivalent transferable model has been developed yet for the European context. A study by Zidek et al. (2003) developed the pCNEM (clear definition for acronym not given) model following principles of the SHEDS model and applied it in London. MEs used for the pCNEM model were however calculated predominantly with data collected in North America and the application for London was not validated by monitoring data.

<i>Study</i>	<i>Pollutant</i>	<i>Study Location</i>	<i>Ambient Model</i>	<i>Spatial Precision</i>	<i>Transferability</i>	<i>Individual/Population Level</i>	<i>Validation</i>
AB²C (Dons, Van Poppel, Kochan, et al. 2014)	BC	Flanders, Belgium	LUR, adjusted to hourly	2386 (sub)zones of Flanders	Within Flanders	Population (individual possible)	Yes
Borrego et al. (2006)	PM ₁₀	Lisbon, Portugal	Hybrid model	2km population grid	No	Population	No
Borrego et al. (2009)	PM ₁₀ (O ₃)	Portugal	air quality - exposure integrated forecasting system (MM5-CHIMERE)	10km population grid	No	Population	No
EXPAH (Gariazzo et al. 2014)	PM _{2.5} , (PAH)	Rome, Italy	Chemical transport model	Residential areas	Yes	Individual (scaled to city level)	Yes
EXPAND (Baklanov et al. 2007; Hänninen 2005b; Kousa, Kukkonen, et al. 2002)	PM _{2.5} (NO ₂)	Helsinki, Finland	Dispersion	100m population grid	No	Population	No
EXPOLIS (Kruize et al. 2003; Hänninen et al. 2003)	Not specified	Athens (Greece), Basel (Switzerland), Helsinki (Finland), and Prague (Czech Republic)	None	None	Yes, in areas where monitored data of study areas is representative	Population (individual possible)	Comparison with other monitoring results
Gerharz et al. (2009)	PM _{2.5} , PM ₁₀	Munster, Germany	Dispersion with temporal adjustments	Individual level time-space location	Yes	Individual	No
STEMS (Briggs et al. 2005; Gulliver & Briggs 2005)	PM ₁₀	Northampton, UK	Dispersion	Individual level time-space location	Yes	Individual	Yes, for model components

Table 4: European studies which developed integrated personal exposure models using the microenvironmental approach. PAH = Polycyclic aromatic hydrocarbons

A number of European studies have developed personal exposure models including MEs, these are presented in Table 4. The largest of these studies was the EXPOLIS (Air Pollution Exposure Distributions of Adult Urban Populations in Europe) study. It used a probabilistic exposure modelling framework based on collected information from 1427 people in 7 European cities. It was aimed to compare exposure of different scenarios or population groups. In order to estimate concentrations for different MEs, concentration distributions were sampled for each ME, either based on monitored or modelled concentrations. The modelling framework has been used in several applications (Kruize et al. 2003; Hänninen et al. 2003). Only crude estimates for transport MEs were included in the framework (sampled from concentration distributions at roadside stations, or using home indoor estimates). The model also did not include geographical information, apart from distinctions of inter-country or urban-rural differences (Kruize et al. 2003; Hänninen et al. 2003; Hänninen et al. 2004; Schweizer et al. 2007; Kousa, Oglesby, et al. 2002).

The Helsinki based EXPAND (Exposure to air pollution, especially to nitrogen dioxide and particulate matter) model combines a GIS-based PM dispersion model (based on the principle that PM disperses away from sources) and an ME model which takes into account population-level time-activity data and trips. The ME component of the model however only included one ratio for indoor to outdoor differences, and the approach does not follow an individual's time-activity. It can therefore not be used to describe variability between personal exposures (Baklanov et al. 2007; Hänninen 2005b; Kousa, Kukkonen, et al. 2002). The EXPAND model has later been applied as part of the European FUMAPEX (Integrated systems for forecasting urban meteorology, air pollution and population exposure) study in Oslo, Copenhagen, and Helsinki.

Borrego et al. (2006) developed and applied an integrated model using data from Lisbon. Their approach to ambient pollution uses a hybrid model (linking a meteorological model, a dispersion model, and a fluid dynamic model). Results were displayed in a GIS-based grid (grid accuracy: 2km); population exposure was then modelled per grid cell per time (inhabitants per grid cell per hour). Adjustments for different microenvironments were then applied to the grid cell estimates (Borrego et al. 2006). Borrego et al. (2009) developed a similar exposure modelling approach for Portugal. The model was based on predictions for ambient concentrations from a hybrid modelling system (combining several models) and predicts exposure for 10km grid cells. Simple ratios were applied to adjust for indoor MEs. Both studies (Borrego et al. 2006; Borrego et al. 2009) are specifically aimed at predicting exposure for the population in the study area. Direct transferability of the model to other areas is not possible, as data underlying the developed grid cells, such as number of inhabitants is location specific.

In a study by Gariazzo et al. (2014) the EXPAH (Population Exposure to PAH) model was developed for Rome, Italy. The model uses a chemical transport model (designed to calculate the lifecycle of chemical components in air pollution) to estimate ambient PAH concentrations in a regular grid of 60x60m. Exposure for the elderly and children was assumed to take place mainly in their neighbourhood. All-day exposure was therefore estimated by residential area, modified by ratios for different MEs, such as home or school indoors. Exposure in transport was considered equal to ambient concentrations.

A further modelling system was developed by Gulliver & Briggs (2005) with the integrated STEMS (Space-Time Exposure Modelling) model. Ambient PM was calculated using a dispersion model and combined with microenvironmental model components, as well as time-activity data. Random sampling was used to extract time-activities for virtual individuals, and a GIS based time-space-activity model was developed to simulate these individual's transport routes. Modifications for each ME were applied by inside-to-outside ratios (Gulliver & Briggs 2005); (Briggs et al. 2005).

Dons, Van Poppel, Kochan, et al. (2014) developed the AB²C model combining temporally and spatially refined ambient predictions (using time-adjusted land-use regression, described below) and fixed ratios for adjustment of indoor MEs. Time-activity was calculated using FEATHERS (Forecasting evolutionary activity-travel of households and their environmental repercussions), which also determined destination and duration of trips between subzones (2386 areas in Flanders, Belgium). Adjustments for different MEs were made using ratios. The model was validated using concentrations data collected by 54 volunteers during one week. The Pearson's correlation between modelled and monitored personal exposure for this model was 0.452.

A pilot study by Gerharz et al. (2009) used time-adjusted results from a dispersion model to predict geographic locations of modelled individuals. MEs were adjusted using comparably refined methods, using ratios for different traffic modes and a mass-balance model for indoor MEs. Mass-balance models are based on physical principles underlying outdoor to indoor pollution differences, and are explained in detail below.

In summary, studies have developed a range of different approaches to calculate exposure using integrated personal exposure models. The approaches used however vary significantly. One main difference for the development and application of an approach is whether it is aimed at predictions at individual or population level. Population level predictions cannot and are not aimed at predicting exposure variability between individuals, but can be used for example to compare regional-level air pollution scenarios (Borrego et al. 2009; Hänninen 2005b). Individual level exposure predictions are usually limited to a relatively small number of subjects, due to the model complexity and computer

processing times. Individual level models can however be used to account for and interpret differences in variability between people. Improvements in computer technology and growing availability of individual-level data makes these models increasingly a possibility for larger population size applications (Borrego et al. 2009; Dons, Van Poppel, Kochan, et al. 2014).

Some of the studies are additionally developed specifically for one study area, while other studies focus on building a transferable model. The approaches also differ in the importance they apply to different modelling components, such as precision of geographical locations, importance and treatment of ambient concentrations for the model, number of different MEs, or type of adjustment applied for different MEs (e.g. using a simple ratio or a mass-balance model for indoor MEs).

2.2.3. Population Time-Activity Data and Models

Data on time-activity patterns can be used to improve exposure estimates as they more accurately reflect the actual locations and MEs in which people get exposed during a day. Exposure assessments without time-activity data may be subject to misclassification especially for the more active part of a population. There are many different ways to include time-activity information in exposure assessment studies. In this section an overview of the most influential types of models is given and study examples are shown.

Most exposure risk assessment studies derive time-activity from regional, national surveys and/or from census data (Fleming & Spellerberg 1999). In order to study and compare population with similar time-activities, groups are commonly selected by age, sex, and location but sometimes also for more complex lifestyle factors (Dimitroulopoulou et al. 2001; Freijer et al. 1998; Hänninen, Karppinen, et al. 2005; Beckx, Int Panis, Uljee, et al. 2009).

The most precise time-activity information of a population is collected by following individuals from the study population through time and space, collecting purpose-designed diary data. Recording time-activity data for the whole study population in an exposure assessment is however often difficult and restricted by limited resources. Diary data is frequently used in combination with personal exposure monitoring to assess the relationships between different time-activities and exposure, but this time-consuming approach is seldom used for large groups (Briggs et al. 2003; Leech et al. 1999).

One way to include time-activity from a large dataset is to use the average time spent for one activity (or in one environment), usually during one day, summarized either for the whole population or for population sub-groups. One example for this method is a study by Dimitroulopoulou et al.

(2001a); in which data from the BBC Survey of Daily Life 1983/1984 is used as the average time spent in different locations for three population sub-groups (homemakers, schoolchildren and office workers). In a study for Lisbon, Borrego et al. (2006) analysed various available datasets and extracted the hourly average number of persons in each microenvironment (residence, office, school, in vehicle). This method is based on a deterministic model in which every set of variables is uniquely determined by parameters in the model.

Probabilistic models work with distributions of different time-activities for certain time periods. Surveys on time-activities can for example provide information about the percentage of people attending certain activities for each hour of the day. These activities can then be probabilistically sampled for a population. As part of the EXPOLIS study, time-activity was entered as the distribution of time fractions spent in different microenvironments (outdoors, home indoors, other indoors, in transport) separately calculated for four population groups (children, adults – working or studying, adults – not working or studying, elderly). These distributions were then sampled using the Latin Hypercube sampling technique. Compared to random sampling Latin Hypercube technique provides a more stable method, based on the division of input variable distributions into ranges prior to sampling (Setton 2007). For the EXPOLIS study, data on time-activity was derived from a time-activity survey in the Netherlands undertaken by Intomart (Kruize et al. 2003; Hänninen 2005a). Similar techniques had been previously applied for a study by the US EPA (Burke et al. 2001). Another example is a study by Zidek et al. (2003) in which a probabilistic model, pCNEM, is presented which estimates personal exposure from large activity surveys.

Research into improving mathematical and software-based probabilistic models is ongoing, and several highly complex models have been developed during the last decade. In principle they usually combine random sampling of existing databases and probability distributions, with other statistical or computational components. De Nazelle & Serre (2006) for example included individual uncertainties and population variation using Monte Carlo simulation. The ALBATROSS (a Learning-Based Transportation Oriented Simulation System) model - developed in 2000 for the Dutch Ministry of Transportation, Public Works and Water Management – was intended as a transport demand model and includes dynamic constraints on schedule decisions. Most of these studies however focused on model development. The ALBATROSS model is one of the few of these approaches which has been applied to real data (Beckx, Int Panis, Uljee, et al. 2009).

In exposure assessment, combining time-activity information with the spatial dimension should always be a high priority, as different locations can cause significant changes to exposure. It is often difficult however to acquire time-activity data that includes full spatial references, as time-activity surveys are usually conducted with sparse spatial information.

2.3. Models for Predictions of Ambient Particle Concentrations

2.3.1. Introduction

As described above, studies have shown that increased exposure to ambient particle concentrations causes adverse health effects in a population. Estimates of ambient particle concentrations are therefore crucial to exposure studies. The most basic exposure estimate is directly derived from monitored ambient concentrations, often based on one or several stationary monitors in the area. Only limited or no spatial variation is included by these approaches (Özkaynak et al. 2013; Avery et al. 2010; Bell et al. 2004). Directly using monitored ambient concentrations to approximate concentration at other locations has as such obvious limitations; a variety of modelling approaches have therefore been developed.

These approaches traditionally included for example stochastic models, which are based on statistical or semi-empirical techniques. These techniques are then used to calculate trends, periodicity and relationships between air pollutants and atmospheric changes. Examples of stochastic models are time-series analyses and frequency distribution analyses. These models work particularly well when predicting short-term trends and are as such useful for short-term forecasting (Karousos 2006).

Improvements in modelling spatial pattern of pollution have been greatly helped by the development of computing and GIS (Geographic Information Systems) during the last few decades. GIS can be used to visualise spatial data, which along with data manipulation and analysis tools makes it easier to detect spatial pattern. It also enables the combination of spatially detailed pollution datasets with explanatory data, and has increased the options for more complex analyses and modelling techniques. GIS-based methods have been used in various exposure studies linked to epidemiological analyses and health risk assessment (Briggs 2005; Ward & Wartenberg 2006; Bellander et al. 2001). The focus of these methods is traditionally on the spatial distribution of ambient pollution. Several types of GIS based methods have been used to predict ambient particle concentrations, such as interpolation or proximity based methods, and dispersion or land-use regression models. In the following paragraphs these models are introduced.

Interpolation methods model the surface between monitoring stations; some of the most common ones include inverse distance weighting, splines and Thiessen triangulation, which are based on deterministic or geometric algorithms. Another interpolation method is kriging (Gaussian process regression), which categorises variation into broad scale trend, local (spatially-structured) variation,

and non-spatial random variation and predicts the area between stations accordingly (Beelen et al. 2009; Jerrett et al. 2005; Briggs 2005).

Another group of models estimates pollution concentrations based on the proximity to source. The underlying assumption of these models is that people living close to a pollution source have a higher exposure than people who live further away, as measured by the distance from the source. One of the more complex proximity models is the focalsum approach; it calculates air pollution levels from information about sources around each cell of a continuous grid. A window of pre-defined shape, weight (depending on source proximity) and size moves over the grid, cell-by-cell, and calculates concentrations of each cell from emission information in the window and from surrounding windows (Loibl & Orthofer 2001; Vienneau et al. 2009).

2.3.2. Dispersion Models

Dispersion models use a technique based on proximity to particle sources, generally relying on the Gaussian plume equation for the dispersion of particles away from each source. Most dispersion models use a combination of pollution data, meteorological data and topographical data. A major advantage of dispersion models compared to most other modelling techniques of ambient air pollution concentrations is that they not only include spatial, but also temporal variations, which makes them some of the most widely applied models for air pollution predictions (Hoek, Beelen, et al. 2008; Baklanov et al. 2007; Daly & Zannetti 2007). Temporal variation is added to the model as part of meteorological data. Meteorological data is generally collected for short time periods (e.g. hours), the temporal resolution of the model can be calculated to the same high resolution. Temporal variability is in addition sometimes added to source related input data, for example data on traffic flow differences throughout the day.

The development of most dispersion models is highly complex. For many areas standard models have been developed and can be used in future applications using purpose designed software (Stocker et al. 2011; Daly & Zannetti 2007). Requirements for input data of these models however can be high including meteorological data, as well as data about air pollution sources (road networks, traffic flow, location and emissions of relevant industrial sites etc.) (Briggs 2005). While meteorological data is usually available for an area, information on sources is not always available. An additional limitation is, that the assumption of the Gaussian plume for the dispersion of particles is not fully accurate (Briggs 2005; Briggs et al. 2003; Gomez-Perales et al. 2004).

2.3.3. Land-Use Regression (LUR) Models

Another commonly applied method to predict ambient particle concentrations is the land-use regression (LUR) modelling technique, which has been increasingly used during the last decade. LUR modelling was firstly introduced by Briggs et al. (Briggs et al. 1997) for the SAVIAH (Small Area Variations in Air quality and Health) study. Since then many studies have successfully adopted the approach (Hoek, Beelen, et al. 2008). LUR modelling is based on the principle that long-term air pollution concentrations at a location can be modelled using information from geographical predictor variables for the same location. Significant predictor variables include for example various traffic representations, altitude, population density, and information on land-use (such as area of parks or built environment) within a certain distances (in buffers) from a location. LUR models are calculated by regressing candidate predictor variables against monitored concentrations for a set of locations (usually between 20 and 100 monitored sites). With the resulting LUR equation, other locations can then be predicted. LUR models are traditionally designed to calculate long-term averages, such as annual concentrations with high spatial accuracy. LUR models have successfully predicted PM concentrations in a variety of locations in Europe and abroad, for example for 20 different study areas in Europe by the ESCAPE study (de Hoogh et al. 2013; Eeftens, Beelen, de Hoogh, et al. 2012) or for Los Angeles, USA by Moore et al. (2007). For urban areas their long-term predictions have been shown to predict better or equivalent to other modelling approaches, such as kriging or dispersion models (Hoek, Beelen, et al. 2008). These comparisons have however been based on R^2 results of LOOCV (leave-one-out cross-validation). A study by Wang et al. (2013), of which the doctoral candidate is co-author, however showed that LOOCV R^2 of LUR models can be inflated due to often low numbers of monitored training sites, as well as potentially large number of predictor variables. In terms of data requirements, the input data needed for the development of a LUR model is often more easily available compared to the input into a dispersion model. The need of monitoring data for a set of locations is on the other hand a clear disadvantage (Hoek, Beelen, et al. 2008).

Several studies have applied LUR models to estimate exposures used to study short-term health effects (e.g. pregnancy cohorts or risk of asthma attacks). Most of these studies have used concentrations from independent monitoring stations, which were not used in the LUR model development, to adjust for temporal variability. The approach how these adjustments are calculated however differs widely between studies. Some studies for example use direct concentration variability (e.g. hourly), while others use temporal pattern (e.g. day of week) aggregated over a longer time period (Brauer et al. 2008; Dons, Van Poppel, Kochan, et al. 2014; Johnson et al. 2013; Mölter et al. 2012; Nethery et al. 2008; Pedersen et al. 2013). Temporal adjustments of LUR models

using independent monitoring data are investigated and compared later in this thesis. A few studies have used a more complex approach by including an element of temporal variability directly into the building process of the LUR model, for example by introducing temporally changing variables in the regression analyses (Noth et al. 2011; Maynard et al. 2007). Data required for the development of such a model are however often not readily available (e.g. road traffic counts with high spatial and temporal frequency) and the incorporation of both spatial and temporal changes into the model can be highly complex. This thesis focuses therefore on the performance of short-term temporal adjustments to an existing LUR model.

2.4. Indoor Microenvironments

2.4.1. Introduction

People in European cities spend on average about 93% of their time indoors (Schweizer et al. 2007), the study of exposure in indoor MEs is therefore of particular importance.

The main influences on indoor particle concentration can be attributed to a few main factors (see Figure 3): penetration of outdoor air through openings and by infiltration, indoor sources (such as smoking cooking, or cleaning), and physical factors, such as deposition and re-suspension. These factors however are themselves variable. Homes for example vary by their built and interior fittings, which in turn influences infiltration, as well as factors such as deposition and air flow. Over time, seasonal factors may be important in connection with heating and window openings. Additionally, the occupant's behaviour has a major influence on indoor particle concentrations. Occupants determine the amount of indoor source activity, as well as frequency of window openings. They are themselves usually also the main cause of resuspension. Another main consideration in terms of indoor particles is the particle size fraction. Smaller particles can for example filtrate more efficiently through the home's wall. Larger particles on the other hand show higher re-suspension in connection to human activities (Hussein et al. 2006; M Toivola et al. 2004). The interaction of all influences on particle concentrations in home MEs is highly complex (Gerharz et al. 2009; Dimitroulopoulou et al. 2006; Hänninen et al. 2004). Hänninen et al. (2004) found that even after adjusting for the known influences on indoor concentrations, a large part of indoor PM remained unexplained.

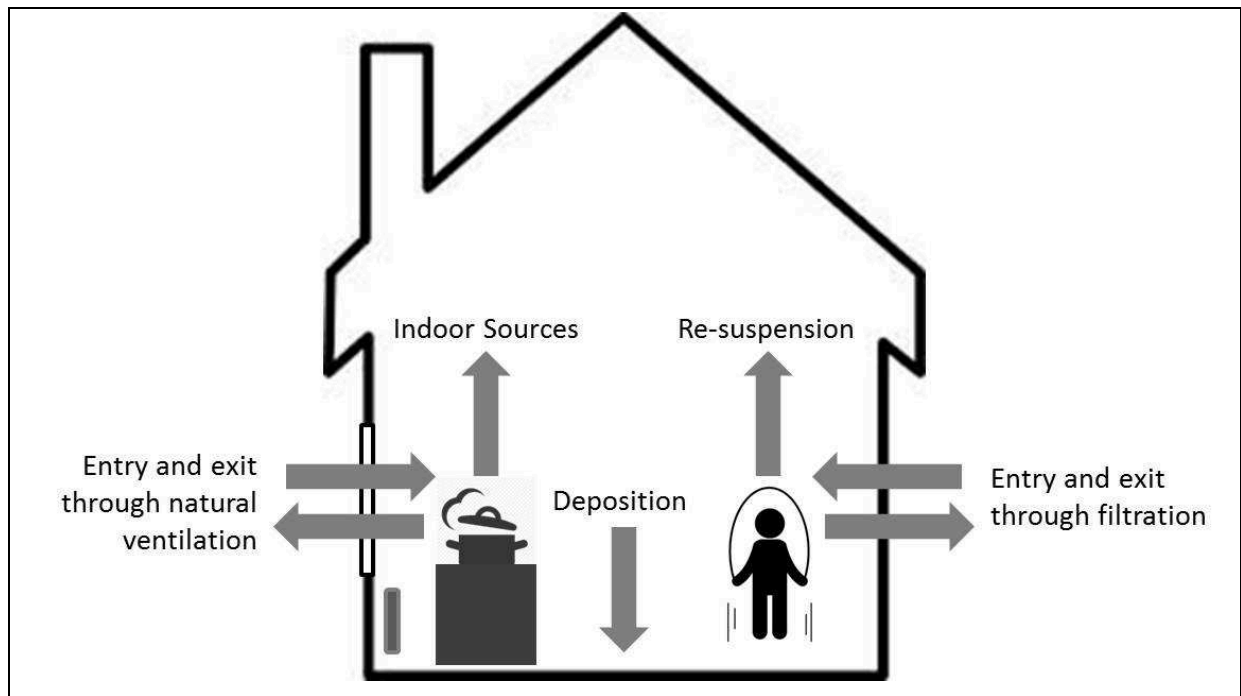


Figure 3: Schematic view of a house with main parameters determining indoor particle concentrations. Source of cooking pot pictogram: Iconfinder (https://www.iconfinder.com/icons/112321/cooking_eating_kitchen_utensils_pan_pictogram_raw_restaurant_simple_icon, accessed: 17th April 2014); Source of rope jumping pictogram: Pictogram Free (<http://pictogram-free.com/46-play/188-play.html>, accessed: 17th April 2014).

Indoor cooking activity has been shown to cause some of the highest particle concentrations indoors (Dimitroulopoulou et al. 2006; Jones et al. 2000; Morandi et al. 1988). At the same time cooking activities are some of the most frequently and ubiquitous source activities in homes. Cooking activity as an indoor source is therefore described in more detail in the following paragraph. Studies showing particle concentrations during cooking events are presented in Table 5.

	Number and type of locations	Extent of campaign	Pollutant	Averaging time	Cooking type (number of events)	Mean	Median
Buonanno et al. 2009	1 laboratory	Min. 3 cooking test each experiment	PM ₂ calc.	1min	Cheese	9,500	
					Wurstel	10,000	
					Bacon	12,000	
					Eggplant	520	
					Bacon, gas stove	5.2	
Buonanno et al. 2010	15 pizzerias	14 month trial time	PM _{2.5}		Pizza Oven	95	
			PM ₁₀		Pizza Oven	123	
Glytos et al. 2010	1 laboratory		PM _{2.5}	1min	Hot plate		
					Onion Frying		
He et al. 2005	15 homes	> 48h each	PM _{2.5}	30sec	Cooking (24)		37
					Cooking pizza(1)		735
					Frying (4)		745
					Grilling (6)		718
					Kettle (25)		
					Microwave (18)		16
					Oven (6)		24
					Stove (4)		57
					Toasting (18)		35
Olsen & Burke 2006	37 homes	7 days each	PM _{2.5}	1min	All cooking types (411)	36	12
					Burned food (6)	470	231
					Grilling (3)	173	155
					Frying (105)	60	33
					Toaster oven (7)	51	45
					Stovetop (40)	17	6.9
					Microwave (20)	11	7.4
					Oven (38)	10	7.7
					Multiple events (43)	29	10
					All electric (141)	43	18
					All gas (42)	29	17

Table 5: Studies on particle concentrations for different types of cooking. Mean and median concentrations are given in $\mu\text{g}/\text{m}^3$. Calc = calculated.

The table shows a wide range of results between studies. Median PM_{2.5} concentrations during cooking for example range from 6.9 $\mu\text{g}/\text{m}^3$ to 745 $\mu\text{g}/\text{m}^3$. The presented results clearly show that different types of cooking, such as use of microwave, grilling, oven use, etc. have a major impact on cooking emissions. Microwave use for example shows both for a study by He et al. (2005), as well as for a study by Olson & Burke (2006) some of the lowest PM_{2.5} concentrations (mean and median below 20 $\mu\text{g}/\text{m}^3$). The same two studies both show some of the highest PM_{2.5} concentrations for grilling with a median of 718 $\mu\text{g}/\text{m}^3$ by He et al. (2005) and 155 $\mu\text{g}/\text{m}^3$ by Olson & Burke (2006). The presented studies differ from each other; study differences are for example the examined particle size fraction, tested types of cooking, number of repeats, setting (pizzeria, laboratory, home), and type of monitoring equipment. These differences make further conclusions difficult.

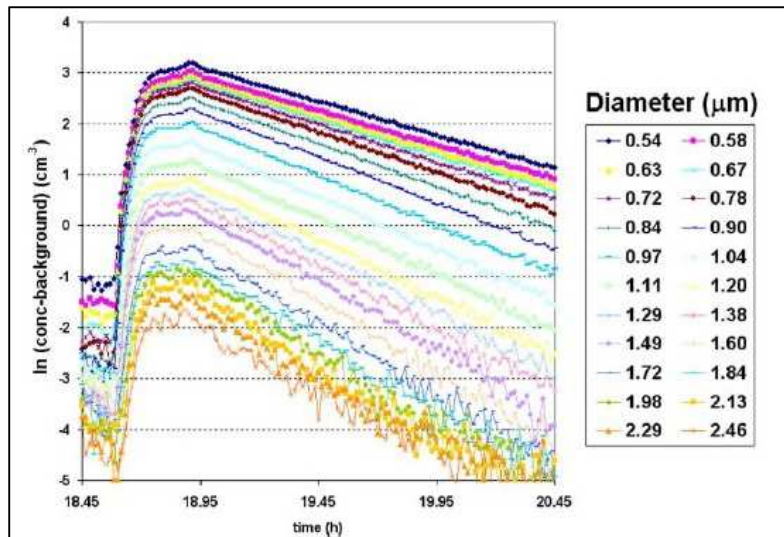


Figure 4: Decay of PM_{2.5} in a home's basement as measured after cooking tortillas in the kitchen. Successive points are 1min apart. Source: Wallace et al. (2004).

In addition to high concentrations during the actual cooking activity, raised concentrations have also been shown for up to several hours after cooking activities finished. Figure 4 shows the decay of PM_{2.5} concentrations by size fraction for the two hours after cooking, as recorded in a study by Wallace (2004).

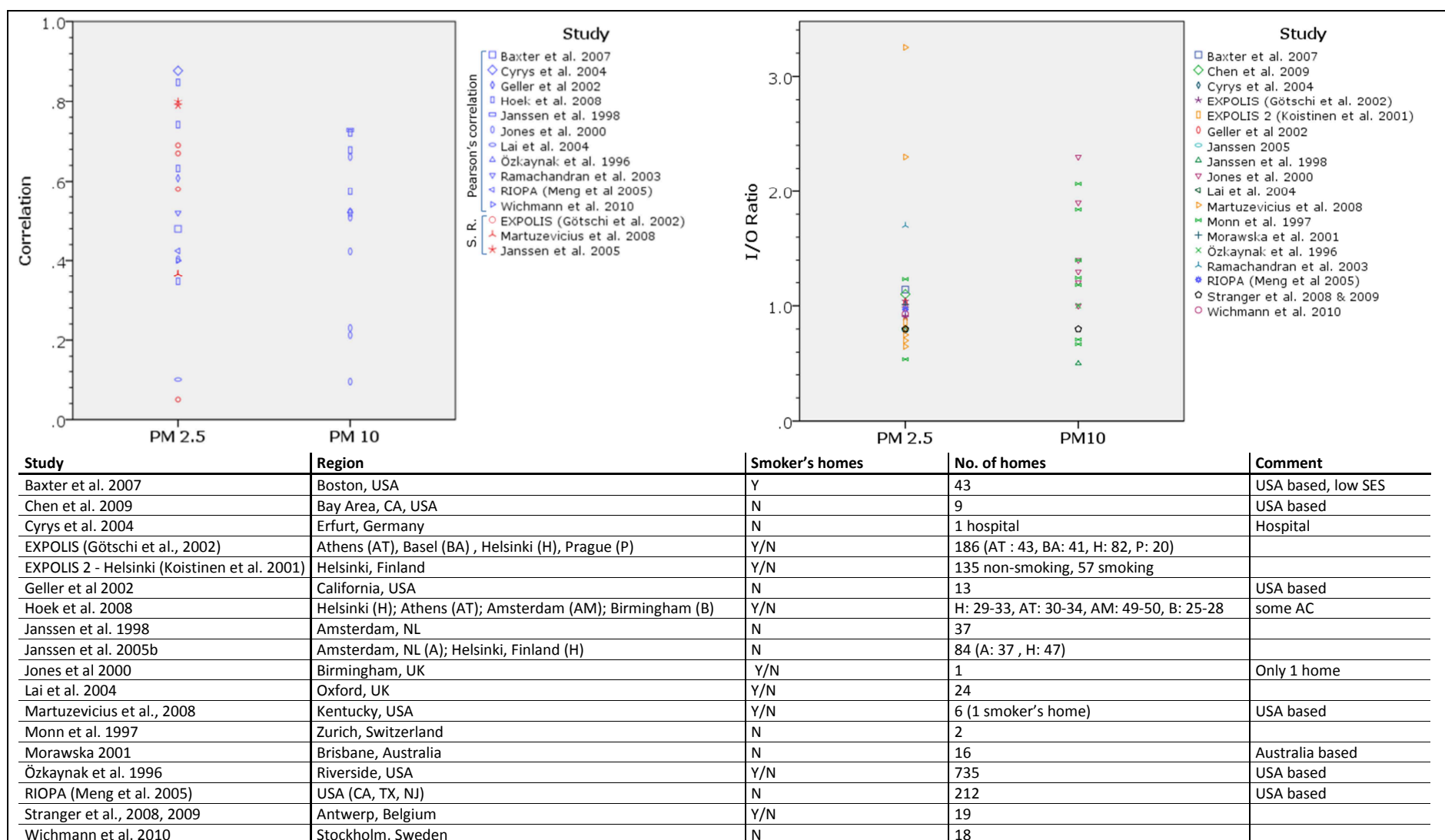
Modelling Indoor Microenvironments

In order to predict home indoor concentrations different types of models have been developed in often highly specialised studies. Important modelling approaches include computational fluid dynamic (CFD) models, mass-balance models, regression models, and simple indoor to outdoor ratios. Indoor CFD models generally focus on the air distribution within the indoor environment, and are therefore especially powerful in explaining PM for indoor spaces with high turbulences or for specialised studies that focus on differences within the indoor environment (Béghein et al. 2005; Chen et al. 2006; Kao et al. 2009). The focus of this thesis is on predicting exposure for the entire home (as part of personal exposure). Differences within the home are considered of minor importance. The CFD approach is therefore here not further investigated. I/O ratios, regression analysis, and mass-balance models have been studied, applied, and compared for this PhD. The three approaches are described in the following paragraphs.

2.4.2. Indoor-Outdoor Ratios

A simple method to get an approximation of indoor PM concentration is to use an indoor-outdoor ratio (I/O ratio), based on the difference between simultaneously monitored indoor and outdoor concentrations. The use of an I/O ratio is based on the idea that despite various additional influences on indoor air, outdoor air is usually a main influence on indoor PM concentrations. Many studies in fact show a significant correlation of indoor PM and concentrations directly outside. The correlation coefficients between indoor and outdoor concentrations from different studies are presented in Figure 5, upper left side. The figure shows that indoor to outdoor correlations vary considerably, between 0.01 and 0.88 for $PM_{2.5}$ and between 0.09 and 0.73 for PM_{10} (results for Pearson's correlation and Spearman's Rho, see Figure 5 for details). The I/O ratios of the presented studies (Figure 5, upper right side) also show a wide range of results. I/O ratios range from 0.45 to 3.25 for $PM_{2.5}$ and from 0.5 to 2.3 for PM_{10} .

Some of the differences between the results can be attributed to varying study set up, intent, and size. Several of these studies for example included smoking, or only studied concentrations at a few houses, which makes direct comparisons between studies difficult (Figure 5). In addition, studies in non-European settings have been included in the summary. The differences in housing material, build, use of heating and air conditioning, as well as behavioural differences (e.g. for cooking) may have caused additional differences in comparison to European studies.



2.4.3. Regression Models

Regression modelling is a technique that seeks to explain a dependent variable using one or more explanatory variables. Regression modelling can be used to develop predictive models of indoor air pollution, but only few studies have applied this method so far (see Table 6). The existing regression models for indoor pollution concentrations vary substantially in their aims and complexity.

Study / Reference	PM size	Model structure	R ²
Harvard Six-City Study: Dockery & Spangler (1981)	Sulphate	$0.04 + 0.75 \cdot \text{Outdoor concentration} - 0.47 \cdot (\text{Central air-conditioning} \cdot \text{Outdoor concentration}) + 0.006 \cdot \text{No. of cigarette per day} + 0.046 \cdot (\text{Central air-conditioning} \cdot \text{No. of cigarette per day}) + 1.08 \cdot \text{Gas cooking} - 0.97 \cdot (\text{Central air-conditioning} \cdot \text{Gas cooking}) + 1.73 \cdot \text{Central air-conditioning}$	0.79
	Respirable	$15.02 + 0.7 \cdot \text{Outdoor concentration} - 0.39 \cdot (\text{Central air-conditioning} \cdot \text{Outdoor concentration}) + 0.88 \cdot \text{No. of cigarette per day} + 1.23 \cdot (\text{Central air-conditioning} \cdot \text{No. of cigarette per day}) - 2.39 \cdot \text{Central air-conditioning}$	0.68
Pilot study, EPA PTEAM study: Wallace (1996)	PM _{2.5}	Daytime: $18 + 0.47 \cdot \text{Outdoor concentration}$ Night-time: $24 + 0.23 \cdot \text{Outdoor concentration}$	0.30 0.05
	PM ₁₀	Daytime: $36 + 0.44 \cdot \text{Outdoor concentration}$ Night-time: $44 + 0.14 \cdot \text{Outdoor concentration}$	0.17 0.01
EPA PTEAM study: Özkaynak et al. (1996a)	PM _{2.5}	$10 + 0.67 \cdot \text{Outdoor concentration}$	0.48
		Daytime: $10 + 0.81 \cdot \text{Outdoor concentration}$	0.49
		Night-time: $9 + 0.56 \cdot \text{Outdoor concentration}$	0.55
	PM ₁₀	$32 + 0.54 \cdot \text{Outdoor concentration}$	0.27
		Daytime: $51 + 0.49 \cdot \text{Outdoor concentration}$	0.20
		Night-time: $20 + 0.51 \cdot \text{Outdoor concentration}$	0.41
	Sulphur (PM _{2.5})	$0.046 + 0.83 \cdot \text{Outdoor concentration}$	0.91
		Daytime: $0.043 + 0.85 \cdot \text{Outdoor concentration}$ Night-time: $0.060 + 0.80 \cdot \text{Outdoor concentration}$	0.93 0.89
	Sulphur (PM ₁₀)	$0.22 + 0.76 \cdot \text{Outdoor concentration}$ Daytime: $0.27 + 0.80 \cdot \text{Outdoor concentration}$ Night-time: $0.19 + 0.72 \cdot \text{Outdoor concentration}$	0.86 0.88 0.84
EPA PTEAM study: Özkaynak et al. (1996b)	PM _{2.5}	$0.64 \cdot \text{Outdoor concentration} + 28 \cdot \text{Smoking} + 9.4 \cdot \text{Cooking}$	0.55
		Daytime: $21 + 0.71 \cdot \text{Outdoor concentration} + 27 \cdot \text{Smoking} + 13 \cdot \text{Cooking} - 2.0 \cdot \text{House volume}$	0.53
		Night-time: $0.53 \cdot \text{Outdoor concentration} + 32 \cdot \text{Smoking} + 4.5 \cdot \text{Air exchange}$	0.71
	PM ₁₀	$0.52 \cdot \text{Outdoor concentration} + 37 \cdot \text{Smoking} + 20 \cdot \text{Cooking} + 5.2 \cdot \text{Air exchange}$ Daytime: $57 + 0.66 \cdot \text{Outdoor concentration} + 29 \cdot \text{Smoking} + 26 \cdot \text{Cooking} - 2.7 \cdot \text{House volume}$ Night-time: $0.45 \cdot \text{Outdoor concentration} + 38 \cdot \text{Smoking} + 12 \cdot \text{Cooking} + 12 \cdot \text{Air exchange}$	0.41 0.39 0.58
RIOPA study: Reff et al., 2005	PM _{2.5}	$0.48 \cdot \text{Outdoor concentration} + 9.05$	0.16
		Dwellings with predominantly outdoor sources: $0.57 \cdot \text{Outdoor concentration} + 4.32$	0.63
		Dwellings with predominantly indoor sources: $0.51 \cdot \text{Outdoor concentration} + 10.68$	0.06

Table 6: Examples of regression models of indoor PM concentrations. Source: Jamieson (2012)

Several studies focus on establishing a relationship between indoor and outdoor concentration by only including outdoor concentration as an explanatory variable. This regression approach includes examples from two major USA based studies: the PTEAM (Particle team) study (Özkaynak et al. 1997; Özkaynak et al. 1996), and the RIOPA (Relationship of Indoor, Outdoor and Personal Air) study (Richmond-Bryant & Reff 2012). Similar to I/O ratios these regression functions do not account for the indoor particle variability related to influences indoors. A distinction was however made, for example by the PTEAM study between daytime and night-time, which accounted to a small extent for the occupant's activities during daytime hours.

A few studies have added additional influences on indoor particle concentration (apart from outdoor concentrations) into the regression model. Dockery & Spengler (1981) for example included the existence of central air conditioning, the number of smoked cigarettes and gas cooking (for Sulphate concentrations) in the model. In a second set of regression models calculated as part of the PTEAM study the following variables were included: smoking, cooking, and house volume. Both studies collected particle concentrations using sampling filters, for 12 hours (PTEAM study) and 24 hours (Dockery & Spengler 1981). The different explanatory variables were regressed against concentration samples for these time periods. During such relatively long time periods indoor air is however influenced by a multitude of factors. Some of these influences, such as window openings and cooking, or cooking and people's movement may also occur at the same time. Additionally not all influences can usually be accounted for in the analysis. Quantifying short-term individual influences from long-term measurements in a regression analysis is therefore problematic.

A study by (Jamieson 2012) developed a regression model for indoor particle concentrations based on data collected at one minute resolution, both for indoor and outdoor concentrations, as well as for diary entries on indoor activities. Explanatory variables of the model include outdoor concentrations, indoor activities (sleeping, standing, walking, cooking), and added time-cycles (for days of week and hours of day). Much of indoor variability in a home takes place on a sub-daily level; this detailed approach therefore promises to account for a larger part of particle concentration variability within homes compared to the regression models presented above. The model by (Jamieson 2012) has been applied in this thesis and compared to other indoor models. A more detailed introduction to this model is given in chapter 5.

2.4.4. Mass-balance Models

Mass-balance (MB) models are based on physical parameters influencing the indoor particle concentration. Several versions of MB models exist, ranging from simple models of air in-filtration to

complex multi-compartment models. In the development of MB models, physical parameters of input and loss to indoor particles have been established. Many studies have looked at single parameters, such as rate of deposition indoors or properties of infiltration of air from the outside (Hänninen et al. 2004). Studies which applied mass-balance models have generally used at least some parameters which were previously established in the literature (Gerharz et al. 2009; Dimitroulopoulou et al. 2006; Hänninen et al. 2004). In the following section two versions of the mass-balance equation are presented and its parameters described. Both models are applied later in this thesis and comparisons between methods for predicting indoor particle concentration are made. The presentation of these two models is followed by a short overview of other types of mass-balance models.

Parameters

In mass-balance models several main parameters describe the physical properties of indoor particle concentrations. The parameters are:

- Penetration efficiency (p),
- Air exchange rate (α),
- Deposition rate (D) (or Decay rate, ' k '),
- Resuspension (R),
- Indoor Sources (S).

The parameters follow the basic influences on indoor particle concentrations, as presented in Figure 3, with penetration efficiency and air exchange rate describing properties of filtration and ventilation between outdoor and indoor particle concentrations:

The penetration efficiency (p) is dimensionless and represents a relation of particle exchange between the indoor and outdoor environments, including infiltration and loss. It can be calculated from a particle decay rate (defined below) and an I/O ratio under stable indoor conditions without any indoor sources present (for example when residents are asleep). The penetration efficiency is dependent on many different influences. Apart from seasonal differences these include for example window ventilation, built of the house, and particle size (Kruize et al. 2003; Thatcher & Layton 1995; Long et al. 2001).

The air exchange rate (α) is a measure of airflow between indoors and outdoors. It is generally calculated in units of airflow per hour.

A number of studies have been looking at the deposition (D) rate of particles in an indoor environment. In order to get a good estimate for particle deposition, a parameter for deposition velocity is taken, generally measured in m/s. The result is then multiplied by the amount of indoor particles and a parameter for the surface area of the microenvironment (or surface to volume ratio), which is then summarized as deposition per hour. Deposition is naturally highly dependent on the amount of activity and air movement in the ME (Dimitroulopoulou et al. 2006). In a few studies the term particle decay rate (k) is used instead of D, which more distinctly includes different types of loss, such as: deposition, adsorption, absorption etc. (Özkaynak et al. 1996). D as applied in this thesis is used synonymous to decay, describing any kind of indoor particle decay.

Infiltration Factors (F_{inf}), is referred to as a parameter by some studies referred to as a parameter. Infiltration factors are simple MB models built from three parameters: p, α , and D, as well as the outdoor pollution (C_{out}).

Some studies also include a parameter for particle resuspension (R) (Thatcher & Layton 1995). Most studies however consider resuspension part of the indoor sources.

Parameters of indoor sources are usually calculated as source emission (in mg/m³) per minute. Only a limited number of source types have been calculated as parameters. Cooking and cigarette source emissions of indoor particle concentrations have been calculated by a few studies (He et al. 2004; Özkaynak et al. 1997; Hänninen et al. 2004). Few attempts have been made to quantify other known sources, such as the impact of people moving about or cleaning (Gerharz et al. 2009; Kruize et al. 2003; Abt, Catalano, et al. 2000; Thatcher & Layton 1995; Chen & Hildemann 2009).

Infiltration Model

In its most simplistic form, an MB model calculates indoor PM only from concentrations that originated outdoors. This form of MB model is called infiltration model (or infiltration factor). For applications with little data availability an infiltration model provides an alternative to the more commonly applied I/O ratio. Similarly to I/O ratios they only require outdoor concentrations as an input, yet they provide in comparison a much more sophisticated function of particle penetration through the building wall. Only one widely recognised version of the infiltration model exists in the literature, by Hänninen et al. (2004) and can be expressed as follows:

Equation 2: Infiltration Model

$$F_{inf} = \frac{C_{out} * p * \alpha}{\alpha + D}$$

The infiltration model (F_{inf}), in the equation above is calculated from C_{out} : the outdoor particle concentration in $\mu\text{g}/\text{m}^3$, α [h^{-1}]: the air exchange rate per time unit, p : the penetration efficiency factor, and D [h^{-1}]: the particle deposition factor (per time unit). The infiltration model as presented here was applied later in this thesis and compared to other models for indoor particle concentrations.

MB model including source emissions

Many versions of mass- balance models exist, which include source emissions. For several of these studies however the equations only differ in naming and exact definition of parameters (e. g. including decay rate instead of deposition factor, or adding resuspension instead of including it into sources), as well as by varying mathematical transformations of the same equation. One of the most widely recognised MB model is based on Koutrakis et al. (1992) and has been applied by a range of studies: Gerharz et al. (2009); Hoek, Kos, et al. (2008); Hänninen et al. (2004); Ferro et al. (2004); Nazaroff (2004). The model equation, as applied by Gerharz et al. (2009) reads as follows:

Equation 3: Mass-Balance Model

$$C_{in} = \frac{C_{out} * p * \alpha + \frac{\sum_{i=1}^n E_i * t}{V}}{\alpha + D}$$

In Equation 3 indoor concentrations (C_{in}) are calculated from outdoor concentrations (C_{out}), the penetration efficiency (p), the infiltration factor (α), and the deposition (D). Emission related parameters are E_i , an emission rate for the i -th of n indoor particle sources, multiplied by minutes of source activity (original equation only mentioned minute time variant ' t ' in the text; ' t ' has been added here to the equation for easier reading), and V [m^3], the room or house volume. The MB model, as presented in Equation 3, has been applied later in this thesis and was compared to other methods to predict indoor particle concentrations.

The two models presented above represent some of the most transferable MB models, which focus on establishing general indoor particle concentrations dependent to concentrations outside. Several other MB modelling approaches for indoor particle concentrations have been developed, which usually follow a more specific focus. Some studies for example developed multi-compartment MB

models, focusing on the differences between rooms (or compartments) within the indoor ME. The most recognised of these examples was developed as part of the INDAIR study (Dimitroulopoulou et al. 2006). Another approach, applied in the INDAIR study is the incorporation of uncertainty in model parameterization. Dimitroulopoulou et al. (2006) used a probabilistic modelling approach, applying probabilistic sampling of distributions for each parameter of the mass balance equation (Dimitroulopoulou et al. 2006). Further development of MB models includes also the introduction of a temporal factor into the model. A temporal component has been applied in different ways, either as a simple lag time (Abt, Catalano, et al. 2000; Schneider et al. 2004), or as a dynamic model (Gerharz et al. 2009).

In general, MB models have proven to be a useful method for predicting indoor particle concentrations by several studies (Kruize et al. 2003; Hänninen et al. 2004; Jamieson 2012). These approaches, however, have some major limitations. MB models are highly dependent on the parameters used. Several of the applied parameters have usually been calculated by other studies. The conditions (housing stock with specific heating and ventilation, behaviour and number of inhabitants etc.) for which the parameters were calculated vary and often differ in other studies to which the model was applied. For example, many European studies used parameters based on American studies, which might not reflect a European context well. In addition, there is a lack of large studies in which parameters were validated. Additionally, parameters for source emissions have not been calculated by many studies.

2.4.5. Predicting In-Office Exposure

Most studies on models of indoor air pollution are conducted for home indoor environments. However, of the time spent indoors on average 30% is spent at work (Schweizer et al. 2007). It is therefore important for personal exposure estimates to account for time spent in the work ME. Most of the working population in Britain, 83.1% (data for Dec. 2013), are employed in the service industries, in which offices are the main type of work environment (Office of National Statistics 2014). Additionally, office work environments can be found for example as part of administrative work in the industrial sector.

Particle exposure in office work MEs was therefore considered of importance. Specialised studies on exposure to air pollutants in non-office work MEs (often referred to as occupational exposure) have been conducted by many studies. In relation to different industrial processes and related pollutants these are highly industry specific (Blair et al. 2007). The focus of this thesis is on population

exposure. Studying air pollution of specific subgroups of industrial workers was considered beyond the scope of this thesis.

Adjustments for office MEs have been applied by a number of studies on personal exposure models. Several of these studies have adjusted work MEs however by using models and adjustments calculated for home indoor MEs (Dons, Van Poppel, Kochan, et al. 2014; Gariazzo et al. 2014; Gerharz et al. 2009; Borrego et al. 2006; Borrego et al. 2009). This assumes that concentrations in indoor office MEs are governed by similar parameters to home indoor MEs. Modern office buildings however show a range of differences in comparison to most home MEs (see Figure 6). Purpose-built office buildings today usually do not have windows that open and are fully air conditioned (Hänninen, Palonen, et al. 2005). In Europe few homes share these characteristics. Centralised air conditioning systems filter air from the outside before distributing it in the building. Air conditioning has been shown by several studies to reduce concentrations of $PM_{2.5}$ and PM_{10} (Gupta & Cheong 2007; Thornburg et al. 2001; Croxford et al. 2000). In addition, the incoming air is usually drawn in from roof level, where concentrations can be much lower compared to the ground. There is rarely any source activity from cooking in offices and often much less vigorous movement among working people, compared for example from playing children in many homes (Burke et al. 2001; Hänninen, Palonen, et al. 2005).

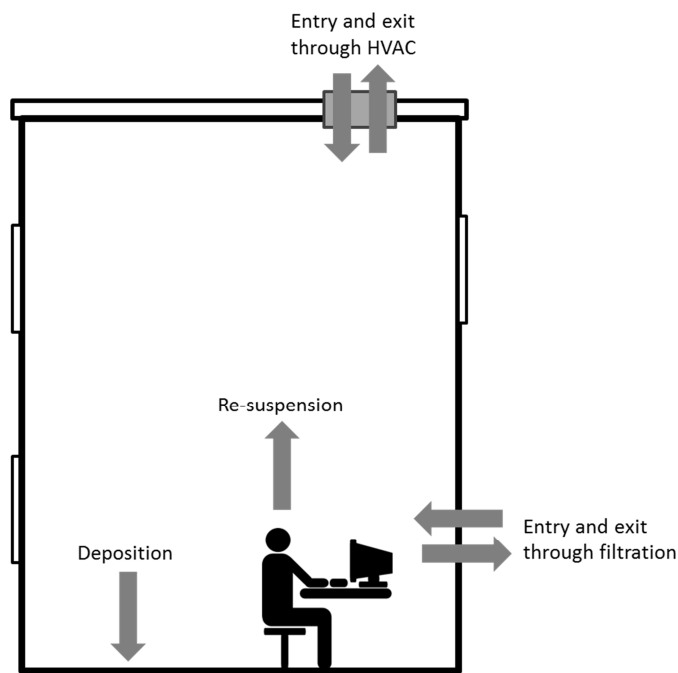


Figure 6: Schematic view of an in-office environment with main parameters determining in-office particle concentrations. Source of office worker pictogram: Clker (<http://www.clker.com/clipart-lan-party-pictogram.html>, accessed: 17th April 2014)

Few studies have been conducted on $PM_{2.5}$ and PM_{10} concentrations in fully air conditioned office MEs, a summary of studies is presented in Table 7. Most of the studies were conducted for few offices. One study for $PM_{2.5}$ by Hänninen, Palonen, et al. (2005) includes 94 offices in Helsinki, Finland. For these offices an infiltration factor for outdoor to indoor particle concentrations was calculated at 0.35. The study however has several limitations, namely that full mechanical ventilation is assumed based on age of the building. Work environments built after 1990 were as such all categorised as mechanically ventilated. In addition, the study included general work environments and the sample may include non-office work environments. Two other, smaller studies have calculated I/O ratios for mechanically ventilated office MEs for $PM_{2.5}$. A study by Ho et al. (2004) calculated an I/O ratio of 0.5 from data of two offices in Hong Kong. A study by Horemans et al. (2008) calculated I/O ratios of between 0.34 and 0.77 for five offices in Antwerp, Belgium. A regression equation to calculate in-office concentrations by 'Zufall et al. (2001)' is referred to by Burke et al. (2001). The original reference could however not be retrieved. The regression equation uses outdoor air as the only predictor variable, its intercept is at 3.6, and the slope is 0.18.

For PM_{10} only two small studies were conducted to establish particle concentrations for fully mechanically ventilated offices. One study by Gupta & Cheong (2007) was conducted in Singapore

for data from two offices. An I/O ratio of 0.46 was calculated from the results. Only one European study was conducted for PM₁₀ by Ní Riain et al. (2003). Using data from two office buildings in London, UK, Ní Riain et al. (2003) calculated I/O ratios of 0.66 and 0.69. The study has however some limitations. For one of the offices the mechanical ventilation was only working intermittently, the other office was only partially mechanically ventilated using a window-mounted fan. In addition, the offices faced a major six-lane road, which could have substantially increased outdoor pollution compared to most other locations in the city and in turn may have altered calculated ratios.

	Study	Country	Building type	Number of buildings	Outdoor to Indoor Adjustment	Limitations
PM _{2.5}	Hänninen, Palonen, et al. (2005)	Finland	Work places	94	0.35*+0.12	All work places after 1990
	Ho et al. (2004)	Hong Kong	Office	2	0.5	
	Horemans et al. (2008)	Belgium	Offices	5	0.34 – 0.77 (mean: 0.45)	
	Burke et al. (2001), as by Zufall et al. (2001)	USA	Offices		Regression: 3.6 + OUT * 0.18	
PM ₁₀	Gupta & Cheong (2007)	Singapore	Offices	2	0.46	
	Ní Riain et al. (2003)	Ireland	Offices	2	0.66 - 0.69	Intermittent use of AC, window mounted fans

Table 7: Studies on outdoor to indoor concentration adjustments of PM_{2.5} and PM₁₀ for mechanically ventilated offices.

*Infiltration factor, OUT = outdoor concentrations, AC = air conditioning system

Despite mostly small datasets and other limitations the results from these studies show that particle concentrations in mechanically ventilated offices clearly are lower compared to concentrations outdoors (I/O < 1). For both PM_{2.5} and PM₁₀, the office I/O ratios presented in Table 7 are also lower than most home I/O ratios presented in Figure 5. This indicates that modern office MEs differ from home indoor MEs, and this fact should be reflected in the modelling approach applied for each of these MEs.

2.5. In-Traffic Microenvironments

2.5.1. Introduction

Peak daily exposures are often experienced by people in traffic situations (Kaur et al. 2005b; McNabola et al. 2008; McNabola et al. 2009a). Study of in-transport exposure is therefore important. Variability of particle concentrations in transport is often high and exposure in transport MEs is therefore complex. High variability of exposure in transport MEs is related to several factors:

1) People in traffic are close to many highly temporally and spatially varying sources. 2) People's temporal and spatial positions often change rapidly as well. 3) People may occupy certain transport MEs in traffic such as buses and cars. Exposure inside these vehicles is different from concentrations directly outside.

A schematic view of different positions in traffic in relation to traffic-related particle concentrations is given in Figure 7. The figure shows the position of different transport modes in a road environment in relation to peak particle concentrations around traffic sources. As mentioned before, particle pollution reduces rapidly away from sources, such as vehicles. The impact of this reduction was confirmed for pedestrians in a study by Kaur et al. (2005). The study shows significantly lower PM_{2.5} concentrations for walking volunteers with walking position away from the kerb, compared to kerbside walking positions. In-traffic cyclists have additionally recorded higher particle exposure compared to exposure for pedestrians on the sidewalk (Kaur et al. 2005b; McNabola et al. 2009a). The difference in particle concentration between the kerb and a position on the sidewalk can be referred to as horizontal displacement (Pielke & Uliasz 1998). In Addition to horizontal displacement, vertical displacement from particle source can be important, as concentrations decrease with height. Breathing height for example differs between people. This in turn causes differences in exposure, especially between children and adults. Vertical displacement is of particular importance for air pollution monitoring, as inlets of air pollution monitors at network monitoring stations are usually placed at about 3m height, where particle concentrations are lower than at exhaust level or at breathing heights (Gulliver & Briggs 2007). In addition, monitoring stations along roads (so called roadside stations) are seldom directly located at the kerb, which reduces concentrations due to horizontal displacement.

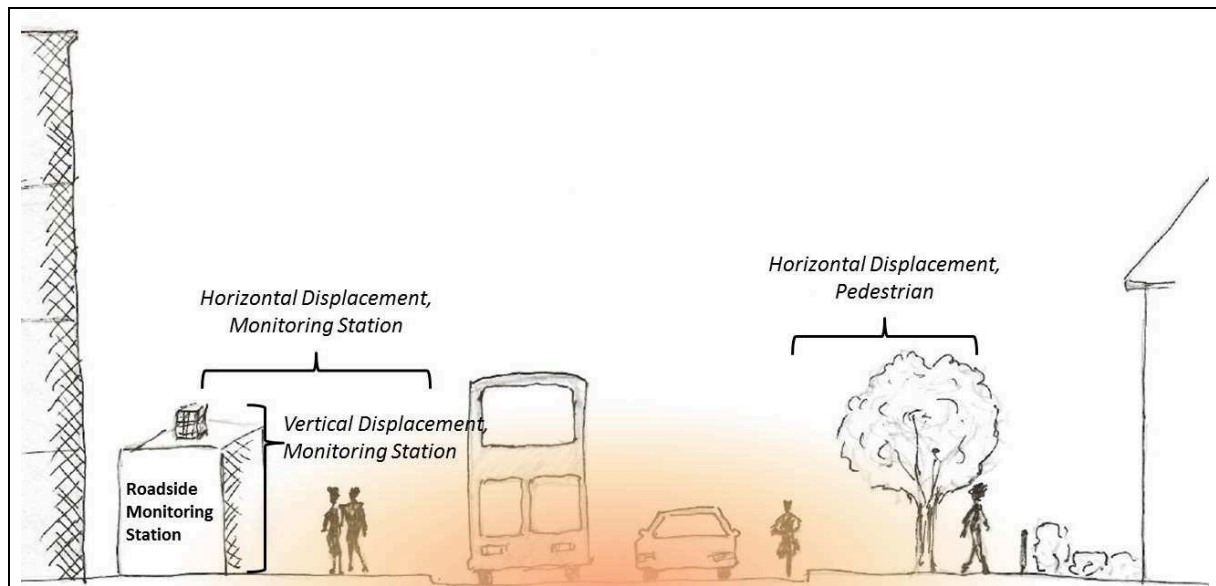


Figure 7: Schematic view of a street cross-section with people and stationary monitors in relation to particle air pollution (peak particle concentration in orange).

In Figure 7, one example of a possible street layout is given. Differences in street configuration can significantly alter the propagation of particles away from road sources. Influences include for example width of road area, width of sidewalk, height and distance of buildings. In addition, wind plays a major role in propagation of particles away from sources. A street canyon refers to roads, which are flanked by tall continuous building fronts on both sides. This canyon situation produces a microclimate with specific conditions for particle pollution, resulting often in high particle concentrations (Elbir et al. 2010; Zhou & Levy 2008; Zwack et al. 2011).

Studies on in-traffic exposure to $PM_{2.5}$ and PM_{10} are presented in Table 8. All of the presented studies collected monitoring data in bespoke transport ME monitoring campaigns. Most of these campaigns are of relatively short (few days) duration and repeatedly record particle concentrations along the same routes using high-frequency monitoring equipment (minute resolution). For several reasons comparisons between these studies are however difficult:

1. The aims and objectives of studies differ. Some studies have been conducted to compare different types of transport modes (Adams et al. 2001; Kaur et al. 2005b; McNabola et al. 2008; McNabola et al. 2009a; Zurbier et al. 2010). Several other studies focus on specific personal exposure, for example for taxi drivers (Pfeifer et al. 1999a). Again, others investigate details of one specific transport mode, such as McNabola, Broderick & Gill (2009), who investigate the effects of inter-vehicle spacing on in-car concentrations.
2. Furthermore the studies vary by
 - a. Investigated mode(s) of transport,

- b. types of route,
- c. geographical setting (for example high traffic or low traffic route),
- d. season and meteorological conditions,
- e. types of monitoring equipment used (for example light scattering or gravimetric method),
- f. Background particle concentration levels during monitoring.

For cycling and walking, proximity to other people, passing smokers in the street and, position on the pavement/road can additionally alter concentrations. For motorised vehicles variation can be caused by the number of passengers, fuel type, vehicle upholstery, use of air conditioning/vents, or whether windows were open or closed.

In the following paragraphs results for the different types of MEs are summarised. Despite the differences between studies, some conclusions can be drawn for the different transport MEs, which are summarised also in the following paragraphs.

Table 8: European studies on in-transport exposure to PM_{2.5} and PM₁₀. Results for different transport modes are marked in colour: walking = blue, bicycle = green, car = orange, bus = red, railway = purple. ¹ Summer, ² Winter, ³ based on median concentrations, GM = geometric mean (otherwise arithmetic mean), RS = roadside monitoring station, BG = background monitoring station, ped = pedestrian, cyc = cyclist

Publication	City	Aim/ Main Objective of Study	Sample No. / Sample Duration / Campaign Duration	Mode	Mean concentration in µg/m ³ (Median)	Ratio with Mon. Station	Ratio with modes
PM_{2.5}							
Adams et al. (2001)	London, UK	Investigating personal particle exposure in transport microenvironments	Study A: 345/ 3-4miles/	Bicycle Bus Car Subway	34.5 (GM) ¹ , 23.5 (GM) ² 39.0 (GM) ¹ , 38.9 (GM) ² 37.7 (GM) ¹ , 33.7 (GM) ² 247.2 (GM) ¹ , 157.3 (GM) ²	About 2x for all modes (BG)	
			Study B: 120/ /	Bicycle	34.2 (GM) ¹		
Asmi et al. (2009)	Helsinki, Finland	Commuter and driver exposure in buses and trams	4d each/ /8d	Bus		0.95 - 2.87 (BG)	
Briggs et al. (2008)	London, UK	Effects of travel mode on particle exposure	46/ 12.8min / 7d 46/ 3.7min/ 7d	Walking Car	6.59 (PM _{2.5} - PM1) 3.01 (PM _{2.5} - PM1)		Ped/car: 2.19 (PM _{2.5} - PM1)
Fondelli et al. (2008)	Florence, Italy	Evaluate exposure inside commuter vehicles	15/ /4d 14/ /4d	Bus Car	56 39	+ 32µg/m ³ (BG) + 20µg/m ³ (BG)	
Gulliver & Briggs (2004)	Northampton, UK	Evaluation of personal exposure to particles in transport microenvironments	74/ /38h	Walking Car	15.06 15.54		
Gulliver & Briggs (2007)	Leicester, UK	Investigation of particle exposure during journeys	66 (2 routes)/ /10d	Walking Car	10.9 (PM _{2.5} - PM1) 8.3 (PM _{2.5} - PM1)		Ped/car: 1.36
Hurley et al. (2004)	London, UK	Assessment of harmfulness of tunnel particle exposure in the London underground	9d	Subway	130 - 200		
Pfeifer et al. (1999)	London, UK	Exposure comparison between taxi drivers and office workers	Car: 10/ 16h/ 7d Subway: 4/ 8h/ 2d	Car Subway	33.36 246		
Kaur et al. (2005a)	London, UK	Pedestrian exposure of air pollution along a busy road	155/ ≥20min/ 12d	Walking	37.7, 33.8 (GM)	2.34 (BG)	
Kaur et al. (2005b)	London, UK	Assessing exposure for different modes at street canyon intersections.	Walking: 56 (2 routes)/ 18min Cycling: 48/ 18min/ Bus: 42/ 18min/ Car1: 29/ 18min/ Car2: 22/ 18min/	Walking Cycling Bus Car 1 Car 2	27.5, 23.8 (GM) 33.5, 30.6 (GM) 34.5, 31.8 (GM) 38.0, 34.9 (GM) 41.5, 39.0 (GM)	approx. 3, for all modes (BG)	
(McNabola et al. 2009a)	Dublin, Ireland	Analysing principle components of air pollutants in four transport modes.	200/3km	Walking Bicycle Car Bus	63.45 88.14 82.73 128.16		
(McNabola et al. 2009b)	Dublin, Ireland	Assessing the influence of inter-vehicle spacing on in-car exposure.	20/ 25min/	Car	103.64		

Publication	City	Aim/ Main Objective of Study	Sample No. / Sample Duration / Campaign Duration	Mode	Mean concentration in $\mu\text{g}/\text{m}^3$ (Median)	Ratio with Mon. Station	Ratio with modes
McNabola et al. (2008)	Dublin, Ireland	Comparison of exposure between modes	44/ 5km/ 48/ 5km/ 45/ 5km/ 46/ 5km/	Car Bicycle Bus Walking	88.95 71.61 103.81 66.27		Car/cyc: 1.49 & 2.85 Car/ped: 1.34 & 1.57 Cyc/ped: 1.41 & 1.75 Bus/ped: 3.09 & 2.14 Bus/cyc: 1.38 & 3.21
Molle et al. (2013)	Paris, France	Exposure inside buses	32/ 40min/ 10d	Bus	59	1.28 (RS)	
Zuurbier et al. (2010)		Assessing the effect of mode choice, fuel type and route on commuter's exposure to air pollutant	10d (buses), 14d (cars), 16d cycling/ / 47d	Diesel Bus Electric Bus Diesel Car Gasoline Car High traffic cycling Low traffic cycling	68.7 (39.1) 40.5 (27.7) 101.3 (59.7) 114.8 (73.6) 72.3 (49.8) 71.7 (65.2)	2.98 ³ (BG) 2.67 ³ (BG) 1.57 ³ (BG) 1.95 ³ (BG) 2.11 ³ (BG) 2.26 ³ (BG)	

PM₁₀

Briggs et al. (2008)	London, UK	Assessing the effect of transport mode on exposure.	46/ 12.8/ 7d 46/ 3.7min/ 7d	Walking Car	37.5 10.7		
GULLIVER & BRIGGS (2007)	Leicester, UK	see PM _{2.5}	66 (2 routes)/ /10d	Walking Car	22.1 15.1	1.67 (RS) 1.34 (RS)	Ped/car: 1.42
Gulliver & Briggs (2004)	Northampton, UK	see PM _{2.5}	74/ / 38h	Walking Car	38.18 43.16		
Praml & Schierl (2000)	Munich, Germany	Assessment of particle exposure in trams and buses in Munich	85/ 4h/ 52/ 4h/ 54/ 4h/	Bus Circular Bus Radial Tram	164.95 (80 - 236) 110.08 (90 - 134) 183.41 (71 - 279)	1.70 – 3.32 (RS) 2.45 – 2.91 (RS) 1.73 – 3.93 (RS)	
Zuurbier et al. (2010)	Arnhem, the Netherlands	Exposure in different commuter transport modes and the effect of fuel type and route on exposure levels	11d (buses), 14d (cars), 15d (cycling)/ / 47d	Diesel Bus Electric Bus Diesel Car Gasoline Car High traffic cycling Low traffic cycling	68.5 (46.7) 53.2 (43.5) 78.5 (49.8) 58.7 (47.9) 38.8 (39.3) 37.2 (37.0)	2.49 ³ (BG) 2.24 ³ (BG) 1.44 ³ (BG) 1.26 ³ (BG) 1.10 ³ (BG) 1.13 ³ (BG)	

2.5.2. Walking

Walking exposure has been studied in comparison to car exposure by three British studies both for $PM_{2.5}$ and PM_{10} . The studies by Gulliver & Briggs (2004), Gulliver & Briggs (2007), and Briggs et al. (2008) have been conducted in Northampton, Leicester, and London respectively. Mean $PM_{2.5}$ concentrations monitored during walking ranged between $6.59\mu g/m^3$ and $15.06\mu g/m^3$, mean PM_{10} exposure for walking ranged between $22.1\mu g/m^3$ and $38.18\mu g/m^3$. Both the study in Leicester and the London study found higher pedestrian exposure compared to simultaneously monitored exposure inside the car (both $PM_{2.5}$ and PM_{10}). The study conducted in Northampton however found higher in-car exposure for both particle size fractions. Two studies by Kaur et al. (2005b; 2005a) monitored pedestrian exposure of $PM_{2.5}$, one focusing on variability within walking exposure along a high-traffic route in London, the other focusing on mode comparisons (with cycling, bus, and car) for street canyon intersections. Results for both studies are with $27.5\mu g/m^3$ (Kaur et al. 2005b) and $37.7\mu g/m^3$ (Kaur et al. 2005a) higher than British studies previously discussed, which may be related to the specific street configurations chosen (high-traffic route, street canyon intersection). Two further studies on walking exposure of $PM_{2.5}$ have been conducted in Dublin along high-traffic routes (McNabola et al. 2008; McNabola et al. 2009a). Both show in comparison high mean $PM_{2.5}$ concentrations with 66.27 (McNabola et al. 2008) and $63.45\mu g/m^3$ (McNabola, Broderick & Gill 2009). Concentrations however are the lowest compared to all other modes monitored for these studies along the same route (bicycle, bus, and car). A reason for the generally high concentrations monitored in the studies conducted in Dublin may be related to the fact that the routes were frequently highly congested. For McNabola et al. (2008) the average walking time was given as 25 to 30 minutes, which is similar and sometimes faster compared to the average journey time in the car of 20 to 45 minutes for the same routes.

2.5.3. Cycling

Cyclists are directly influenced by high ambient particle concentrations in traffic. In addition, due to the increased breathing rate during cycling the particle dose may be higher compared to other modes of transport (Strak et al. 2010). This thesis however focuses on exposure; cyclist's dose is therefore not further investigated. Cycling was monitored in several multi-mode studies for $PM_{2.5}$. The lowest concentrations were monitored by two studies in London with mean concentrations between $23.5\mu g/m^3$ and $33.5\mu g/m^3$ (Kaur et al. 2005b; Adams et al. 2001). The two mode comparison studies by McNabola et al. (McNabola et al. 2008; McNabola, Broderick & Gill 2009) in Dublin show, as for walking, high mean concentrations with $71.61\mu g/m^3$ (McNabola et al. 2008) and

88.14 $\mu\text{g}/\text{m}^3$ (McNabola, Broderick & Gill 2009). Cycling to walking ratios calculated for McNabola et al. (2008) are 1.41 and 1.75 for different routes. A multi-mode monitoring campaign was also conducted by Zuurbier et al. (2010); the different modes were however not monitored simultaneously. Mean $\text{PM}_{2.5}$ exposure for cycling was monitored as 72.3 $\mu\text{g}/\text{m}^3$ along a high traffic route and 71.7 $\mu\text{g}/\text{m}^3$ along a low traffic route. Median ratios to a background station are 2.11 (high traffic) and 2.26 (low traffic). The study by Zuurbier et al. (2010) is the only study which also provides monitoring concentrations for PM_{10} . Mean PM_{10} exposure during cycling was recorded as 38.8 $\mu\text{g}/\text{m}^3$ on a high traffic route and 37.2 $\mu\text{g}/\text{m}^3$ on a low traffic route. The PM_{10} ratio of the cycling exposure to a background monitoring station is 1.10 for the high traffic and 1.13 for the low traffic route. Surprisingly, the mean PM_{10} concentrations lie below the mean $\text{PM}_{2.5}$ concentrations. Potential sampling error of the dataRAM monitor (light-scattering method) used for $\text{PM}_{2.5}$ measurements in relation to relative humidity was mentioned by the study's author. PM_{10} concentrations on the other hand were monitored using a Harvard Impactor (gravimetric method).

2.5.4. Car

In comparison to walking and cycling exposure, in-car exposure is not only related to the highly variable ambient concentrations in traffic, but is additionally altered by the car exterior and degree of ventilation between indoor and outdoor air. Ventilation is on one hand determined by the permeability of the car exterior, which is itself related to other factors, such as type of car, make, and age. On the other hand, ventilation depends on the chosen ventilation settings and window openings during the journey (Chan & Chung 2003; Briggs et al. 2008).

In-car exposure was examined by most studies investigating in-transport exposure. The lowest mean concentrations inside cars have been recorded by a study in London (Briggs et al. 2008) and a study in Leicester (Gulliver & Briggs 2007). For Briggs et al. (2008) mean concentrations of 3.01 for $\text{PM}_{2.5}$ and 10.7 for PM_{10} were recorded. For Gulliver & Briggs (2007) a mean $\text{PM}_{2.5}$ concentration of 8.3 $\mu\text{g}/\text{m}^3$ and a mean PM_{10} concentration of 15.1 $\mu\text{g}/\text{m}^3$ were recorded. As mentioned before these results lie even below the simultaneously monitored walking concentrations along the same route. All other studies which compare in-car exposure to exposure in other modes found higher in-car exposure compared to walking, and in most cases also compared to cycling exposures (Adams et al. 2001; Gulliver & Briggs 2004; Kaur et al. 2005b; McNabola et al. 2008; McNabola et al. 2009a; Zuurbier et al. 2010). By far the highest mean concentration of all studies was found by a study on inter-vehicle spacing in congested traffic conditions (McNabola et al. 2009b) for which an average $\text{PM}_{2.5}$ concentration of 103.64 $\mu\text{g}/\text{m}^3$ was monitored. Results for mean concentrations during in-car

exposure of all remaining studies vary widely between $15.54\mu\text{g}/\text{m}^3$ and $88.95\mu\text{g}/\text{m}^3$ for $\text{PM}_{2.5}$ and between $43.16\mu\text{g}/\text{m}^3$ and $78.5\mu\text{g}/\text{m}^3$ for PM_{10} .

Ratios for in-car to pedestrian differences of $\text{PM}_{2.5}$ were calculated at 1.34 and 1.57 for different routes by McNabola et al. (2008). The same study also calculated in-car to cycling ratios of 1.49 and 2.85 (different routes). Comparisons of in-car exposure with monitoring stations show invariably higher concentrations inside the cars. A study by Fondelli et al. (2008) found $\text{PM}_{2.5}$ concentrations at a background monitoring station to be $20\mu\text{g}/\text{m}^3$ lower than monitored in-car concentrations (in-car mean = $39\mu\text{g}/\text{m}^3$). Zurbier et al. (2010) calculated in-car to background ratios for $\text{PM}_{2.5}$ of 1.57 (Diesel car) and 1.95 (Gasoline car), and for PM_{10} of 1.44 (Diesel car) and 1.26 (Gasoline car). The study by (Gulliver & Briggs 2007) calculated an in-car to a roadside monitoring station of 1.34 for PM_{10} .

2.5.5. Bus

Public transport buses are one of the most complex air pollution MEs and concentrations depend on a multitude of factors. Like cars, ambient particle concentrations penetrate inside through the vehicle exterior and through openings, such as doors and windows. The following factors have been shown or suggested to additionally have an impact on particle concentrations and exposure inside buses:

- Ventilation related influences:
 - Frequent door openings at bus stops (Vijayan & Kumar 2010; Song et al. 2009),
 - Self-pollution (Behrentz et al. 2005; Marshall & Behrentz 2005),
- Passenger related influences:
 - Frequent movement of passengers (Vijayan & Kumar 2010; Song et al. 2009),
 - Position of individuals in the bus (front, back, upper or lower deck) (Kaur et al. 2007; Molle et al. 2013),
 - Bus passenger capacity (Kaur et al. 2007),
- Bus type/design related influences:
 - Vehicle upholstery (Kaur et al. 2007),
 - Bus model (year and design) (Kaur et al. 2007),
 - Bus fuel type (Kaur et al. 2007; Zurbier et al. 2010),
- Traffic related influences:
 - Position of the bus in traffic (Kaur et al. 2007; Vijayan & Kumar 2010),
 - Bus speed (Kaur et al. 2007),

- Bus idling and acceleration (Kaur et al. 2007),
- Type of bus route (bus routes are often following high traffic routes).

Several studies have monitored $PM_{2.5}$ and PM_{10} concentrations inside buses (see Table 8) and results show some of the highest mean exposures across all road transport modes. For $PM_{2.5}$ mean concentrations monitored by different studies vary widely between $34.5\mu g/m^3$ and $128.16\mu g/m^3$. For PM_{10} concentrations vary between $53.2\mu g/m^3$ and $164.95\mu g/m^3$. Three of the studies which compare multiple modes show the highest mean concentrations for in-bus exposure (Adams et al. 2001; McNabola et al. 2008; McNabola et al. 2009a), compared to other road transport modes (such as car and bicycle). Two studies however show higher mean concentrations for in-car compared to in-bus exposure (Kaur et al. 2005b; Zuurbier et al. 2010).

Ratios of in-bus exposure compared to other modes have been calculated for $PM_{2.5}$ by McNabola et al. (2008) with in-bus to pedestrian ratios of 3.09 and 2.14 (different routes) and in-bus to cycling ratios of 1.38 and 3.21 (different routes). Comparisons of in-bus exposure to monitoring stations have been undertaken by several studies. Asmi et al. (2009) shows ratios between 0.95 and 2.87 (for different days and bus ages) of in-bus to background $PM_{2.5}$ concentrations. Fondelli et al. (2008) found in-bus $PM_{2.5}$ concentrations $32\mu g/m^3$ higher than at a background station (mean in-bus concentration = $56\mu g/m^3$). A Parisian study by Molle et al. (2013) shows a ratio between concentrations in-bus and at a roadside monitoring station of 1.28. In-bus to background concentration comparisons have been undertaken by Zuurbier et al. (2010) both for $PM_{2.5}$ and PM_{10} . Ratios for $PM_{2.5}$ are 2.98 for the Diesel bus, and 2.67 for the electric bus, and for PM_{10} 2.49 for the Diesel bus and 2.24 for the electric bus (median ratios).

Some studies have attempted to model the inside of buses. A study based in Toledo, USA by Vijayan & Kumar (2010) for example developed regression models for several small particle size fractions ($PM < 1\mu m$) for which candidate predictor variables were tested for ambient concentrations, as well as for example for door opening status and temperature. The final model for PM_{10} was entirely based on outdoor concentrations and meteorological variables (ambient air visibility, temperature inside and outside the bus, relative humidity inside and outside the bus). The model resulted in an R^2 adjusted of 0.808. Another model, based in York, UK, was developed by Song et al. (2009) for particle fractions between $0.75\mu m$ to $15\mu m$ using a mass-balance approach. Regression of modelled against monitored in-bus concentrations for different size fractions showed R^2 results between 0.392 and 0.709.

2.5.6. Rail

Little is known about particle exposure in different types of railway transport MEs, such as local trains, subways or trams. Railway tracks, such as for subways and local trains are often entirely separated from other road related traffic and as such are not influenced directly by road traffic sources. Railway trains however are themselves sources of particles, for example from brakes or from engines (diesel trains). Studies undertaken in subways have shown that railway related emissions can build up in tunnels and can cause very high concentrations.

Several studies show that particle concentrations in the London subway are several orders of magnitude above levels found in other modes of transport. For $PM_{2.5}$ Adams et al. (2001) reported mean concentrations of $247.2\mu g/m^3$ inside the London subway, compared to between $34.2\mu g/m^3$ and $39.0\mu g/m^3$ for other modes of transport (bicycle, car, bus). Similarly, Pfeifer et al. (1999) found London subway concentrations of $PM_{2.5}$ to be seven times those found in taxis. Hurley et al. (2004) also investigated concentrations in the London subway system and monitored concentrations between $130\mu g/m^3$ and $200\mu g/m^3$. A study by Sitzmann et al. (1999) has however shown for PM_5 that concentrations in London subways have a different chemical composition (e.g. high on Fe/Si-rich particles) and size distribution compared to ambient concentrations (measured for cyclists). In consequence health effects caused by in-subway particles may also be different.

High concentrations in subway systems only occur within tunnels. Large parts of local railway systems are however above ground. For example, 55% of the London tube runs above ground (http://en.wikipedia.org/wiki/London_Underground). A study by Hurley et al. (2004) shows between 4.74 to 10.9 times higher concentrations for times below ground compared to above ground for different lines of the London tube system.

For PM_{10} , only one study has been conducted for concentrations in a railway transport mode by Praml & Schierl (2000) for in-tram concentrations in Munich, Germany. In-tram PM_{10} concentrations by that study range between $71\mu g/m^3$ and $279\mu g/m^3$ and ratios of in-tram to roadside monitoring station are given as between 1.73 and 3.93.

3. Description of Monitoring Campaigns and Data

3.1. Introduction

3.1.1. Monitoring Campaign Overview

The aim of this thesis is to develop a model for personal exposure. For the development, and validation of this model and its components, data of monitored particle concentrations was essential. Concentration data was available from existing fixed-site monitoring stations for some applications. Further monitoring of particle concentration data was however required for personal exposure, for indoor and outdoor concentrations at home, as well as for transport modes. In this chapter the collection of air pollution data during several monitoring campaigns and resulting particle concentration data is described, as well as its preparation for the following analysis.

Different types of monitoring campaigns were undertaken to collect data on particle concentrations (see also Table 9):

- A) A campaign collecting data at outdoor locations in and around London for the development of a model of ambient air pollution,
- B) A campaign that monitored inside and outside homes in order to apply and compare several different indoor models,
- C) A personal monitoring campaign, monitoring the particle levels close to individuals during their daily routine, and
- D) A campaign comparing concentrations in transport modes in preparation to calculate adjustments for personal exposure in different modes of transport.
- E) Additionally, the study monitors used for campaigns B, C, and D (Osiris) were co-located with one another for assurance of data consistency and with monitors from the London Air Quality Network (LAQN) in order to compare concentrations between types of monitors.

Air pollution monitoring for the ambient monitoring campaign was undertaken in London and the Thames Valley, all other monitoring was undertaken in central London. All monitoring for this thesis was undertaken between January 2010 and September 2011. Monitoring time periods for each campaign are presented in Table 9. In the following paragraphs the different campaigns are introduced. An additional analysis is added in Appendix B, comparing data monitored simultaneously for indoor, outdoor and personal concentrations.

Year	2010												2011								
Month	1	2	3	4	5	6	7	8	9	10	11	12	1	2	3	4	5	6	7	8	9
A) Ambient:																					
B) Home Indoor-Outdoor:																					
Home A																					
Home B																					
Home C																					
Home D																					
C) Personal:																					
Individual A																					
Individual B																					
Individual C																					
Individual D																					
D) Transport MEs																					
E) Co-location																					

Table 9: Temporal Overview of the monitoring campaigns undertaken as part of this thesis.

A) Monitoring campaign to collect data for the development of a model for ambient concentrations

The personal exposure models developed in this thesis, work with a baseline model for ambient concentrations. Microenvironmental models and adjustments then use the predicted ambient concentrations as baseline input. For the personal exposure model in this thesis, the baseline ambient model is built using land-use regression (LUR) modelling techniques. The development of a LUR model requires spatially distributed ambient PM levels in order to calibrate the weights of model parameters and validate model predictions.

Data collection for this purpose was undertaken as part of the ESCAPE project. The doctoral candidate worked as part of the ESCAPE study between 2008 and 2011. One of her main responsibilities as part of that work was to undertake a bespoke monitoring campaign in preparation for the development of the LUR model for London and the Thames Valley. Personal responsibilities included: site selection, recruitment and communication with site owners, organisation and undertaking of monitoring campaigns (including deployment and collection of equipment) recording of information during site visits, communication with ESCAPE colleagues about equipment, as well as posting of the samples to the laboratory. The ambient monitoring campaign was undertaken between January and December 2010 during which concentrations for different seasons of year 2010 were collected at 21 different locations.

B) Home indoor-outdoor monitoring campaigns

Several types of models for home indoor microenvironments (MEs) have been developed by other studies (see chapter 2). Only little is known about how different modelling approaches compare. The main reason for collecting home indoor and outdoor concentrations was therefore to provide data that enabled application and comparison of different indoor modelling approaches. Outdoor concentrations were used as input for different models from the literature. Simultaneously collected indoor concentrations were used to compare how well different models predicted monitored concentrations. A second reason for collecting home indoor concentrations was to provide data for personal exposure when individuals were at home. Concentrations collected with the home indoor monitor were incorporated into the dataset for simultaneously collected personal exposure for times the inhabitants spent at home. A third reason for collecting indoor and outdoor concentrations was for parameter development. There is a need for further development of some indoor model parameters, especially for the mass-balance (MB) modelling approach. A requirement for the data with which parameter development was undertaken was independence of the datasets used for predictions. The indoor-outdoor monitoring data used for parameter development was therefore collected in homes B, C, and D; the data for model comparisons was on the other hand collected in home A.

Collected indoor-outdoor datasets and their different applications are summarised in Table 10. Data for application and comparison of different indoor models was collected at one home, home A. Indoor-outdoor concentrations were collected for home A during 39 days. Home indoor and outdoor concentrations have been collected at three other locations: homes B, C, and D, indoor concentrations for these homes are used as part of personal exposure between two and four days. The other days for which indoor and outdoor concentrations were collected at home B, C, and D form a dataset for the development of indoor model parameters.

	Total days collected	Indoor model application, in days	Indoor as part of personal exposure, in days	Days used for parameter development
Home A	39	39	39	0
Home B	10	0	2	8
Home C	7	0	0	7
Home D	8	0	4	4

Table 10: Datasets collected during the indoor-outdoor monitoring campaign.

During the home indoor-outdoor monitoring campaign data has always been collected simultaneously inside and outside at the four residences. At home A data was collected for 39 days

during almost one year from April 2010 to February 2011 (see Table 9). At each of the three other residential locations, consecutive days of monitoring were collected in summer 2011, recording 10 days at home B, 7 at home C, and 8 days at home D. In summary 65 days of indoor-outdoor monitoring data were collected. For the data description below all days of indoor and outdoor concentrations are presented together for each different home, regardless of purpose.

C) Personal Monitoring Campaign

Data collected for personal exposure to particles is applied as part of this thesis to validate the personal exposure model developed in this context and to compare model performance to different other approaches of exposure assessment. The personal exposure monitoring campaign formed in addition part of work conducted for the GENESIS (Generic European Sustainable Information Space for Environment) study, an EU 7th framework project.

A personal monitoring campaign was undertaken for one resident of each of the homes at which the indoor-outdoor monitoring campaign was undertaken. Personal monitoring data were collected for a total of 51 working days (see Table 11). During personal monitoring a monitor was kept close to the individual at all times when not at home. Monitoring concentrations when at home were collected with the indoor monitor of the indoor-outdoor campaign.

The individual living at home A collected data between April 2010 and February 2011 for 43 days. For four of these days failure of the indoor monitor however meant that only concentrations away from home were collected. Three very small samples of personal exposure were collected for individuals living at homes B, C, and D (called individuals B, C, and D). Collections for these individuals took place during consecutive days in summer 2011. Individual B and C each collected two days of personal monitoring data, and individual D collected data during four days. It should be noted that it was initially intended to collect a larger dataset for individuals B, C, and D. Repeated failure of the only portable monitor (Dustmate) however limited the amount of successfully collected data. Due to the much larger dataset, the comparison of different variations of personal exposure models in chapter 6 is undertaken only for individual A. Datasets of individuals B, C, and D are used to test transferability of the model to other individuals.

	Main individual	Individual B	Individual C	Individual D	Total
Number of days	43	2	2	4	51

Table 11: Days collected during the personal exposure monitoring campaign.

For all personal monitoring, diary data was recorded for times at home, noting down different activities (i.e. cleaning, cooking, and sleeping) and number of occupants. During times away from home the time that was spent in each ME was noted in the diary. All diary entries were recorded by noting the start and end of each activity or location (in minutes). A GPS was used to record locations when not at home or at work.

D) Transport Monitoring Campaign

As presented in chapter 2, exposure in traffic often differs from ambient concentrations monitored or modelled for roadsides. Different modes of transport, such as vehicles in addition alter concentrations of personal exposure. For some transport ME, such as cars studies have been conducted to adjust for the difference to modelled roadside concentrations. Only few studies however have been undertaken which calculate an adjustment from roadside (walking) to in-traffic (cycling) exposure, especially for PM₁₀ (see chapter 2). Walking and cycling was therefore monitored simultaneously along two routes (one high traffic route and one low traffic route) in order to calculate adjustment ratios. Exposure inside public transport buses is very complex and few studies have examined in-bus exposure (see chapter 2). A second monitoring campaign for mode comparisons was therefore undertaken during which cycling and in-bus concentrations were monitored simultaneously along two routes (one high traffic route and one low traffic route).

	Simultaneous cycling-walking		Simultaneous cycling-bus	
	Low traffic route	High traffic route	Low traffic route	High traffic route
Number of journeys	10	8	15	17

Table 12: Number of journeys undertaken for the transport monitoring campaign for simultaneous cycling to walking and cycling to bus comparisons along a low traffic and a high traffic route.

Both walking-cycling and bus-cycling campaigns were conducted during two weeks in March 2011 (see Table 9). The cycling-walking campaign was undertaken for 10 journeys along a low traffic route and eight journeys along a high traffic route. The bus-cycling campaign was undertaken for 15 journeys along the low traffic route and 17 journeys along the high traffic route (see Table 12). The data collection for the transport monitoring campaign formed in addition part of work undertaken for the MACC (Monitoring Atmospheric Composition and Climate) study, an EU 7th framework project.

E) Monitor Co-location

Different types of air pollution monitoring devices exist (see chapter 2). For the ambient monitoring campaign gravimetric Harvard Impactor monitors were used, which are considered one of the reference methods (to which other methods are often calibrated). For the indoor-outdoor, personal, and transport campaign light-scattering devices were utilised, which have a short-term resolution. Osiris (Optical Scattering Instantaneous Respirable Dust Indication System) and Dustmate monitors were used, which provide a continuous short-term concentration and can be used stationary as well as for portable monitoring. One Dustmate and three OSIRIS, produced by Turnkey (Turnkey Instruments Ltd., Northwich, UK) were used for this study. Both machines (Osiris and Dustmate) are the same and only differ in their packaging. Both devices are therefore referred to from now on as 'Osiris', Dustmate monitors are not mentioned separately. The Osiris monitors were provided by the Department of Epidemiology and Biostatistics, Imperial College. The monitors record at one second intervals and were set up to store data as one minute averages. This high temporal resolution has the advantage that possible short-term concentration changes can be detected easily and studied in detail to detect patterns or potential problems. In order to assure relative consistency of data collected with the study's monitoring devices, the monitors were co-located with each other during periods of monitoring. Results for Spearman's rho correlation between the monitors during these co-locations are displayed in Appendix C. On two occasions the Spearman's rho was below 0.1 and non-significant. The monitors were on these grounds sent in for repairs and calibration. Any monitoring data that had been collected prior to the repair (since the last co-location) was excluded from the datasets. Data was additionally lost on several occasions because the monitors had for example problems with the connection to the power supply.

In several studies it has been shown that light-scattering monitors may not compare well with monitors that are reference-equivalent, underprediction has for example shown by several studies (Gulliver & Briggs 2004; De Jonge et al. 2008; Walden et al. 2010). Osiris monitors were therefore co-located with EU reference equivalent monitors at two network monitoring stations in London during March and April 2011. A more extensive description of all monitoring devices used in this thesis is added in Appendix A

3.1.2. Introduction to Data Description

In this section an introduction is given into the methods applied to describe collected monitoring data. Data collected during the ambient monitoring campaign was provided after laboratory

analyses, as seasonally adjusted annual averages by colleagues from the ESCAPE study. The data was described using mean, standard deviation (SD), and range.

Data for indoor-outdoor and personal, as well as transport monitoring was collected at a minute interval. Description of the datasets is presented for original minute concentrations for the following three reasons. 1) Minute concentrations provide the opportunity to understand the influence of different activities in detail, for example for peak exposures during cooking or in traffic. This provides important information on the main drivers of increased short-term exposure. 2) Many MEs passed during personal exposure monitoring have been visited only for few minutes each time (e.g. bus stops), presentation at a more temporally aggregated level would have been difficult. 3) Few other studies have collected personal exposure at high-temporal resolution over the course of days. The majority of personal monitoring has been conducted using passive samplers. Passive samplers however collect data for a period of time of at least several hours and cannot provide information on peak exposures (Gauvin et al. 2002; Harrison et al. 2002; Janssen et al. 2005; Koistinen et al. 2001; Lai et al. 2004; Violante et al. 2006; Johannesson et al. 2011; Williams et al. 2008; M Toivola et al. 2004; Adgate et al. 2007; Monn et al. 1997).

Summary statistics for each of the minute datasets are presented below using median and interquartile range (IQR), as main descriptive statistics. These parameters were preferred over the more commonly used mean, and standard deviation (SD), as they are more informative for skewed datasets. All datasets presented here at a minute level are positively skewed. Arithmetic mean and SD are given in brackets as additional information. The 99th percentile (99th P.) and range are given in addition, where applicable.

In some of the following sections of this chapter, traffic data is used to describe local particle sources at the monitored location or along a journey. All traffic data used were provided by the London Atmospheric Emissions Inventory (LAEI). The LAEI calculated traffic volume for the year 2008 as average daily traffic (DAT) counts for all major and medium sized roads in Greater London (London Atmospheric Inventory 2008: 2011-09-15).

In the following sections the different monitoring campaigns and collected data are described, starting with the ambient monitoring campaign, followed by the residential indoor-outdoor monitoring campaign. Then, campaign and data of personal monitoring are described, followed by the transport monitoring campaign and results from monitor co-locations. In the last section, the data is prepared for analyses in chapters 4, 5, and 6. All calculations for data descriptions, as well as for analyses have been undertaken using SPSS software. Maps have been developed using ArcGIS software.

3.2. Ambient Monitoring Campaign

3.2.1. Data Collection

In the following section the site selection, set up and execution of the ambient monitoring campaign is described. A careful selection process was undertaken to prepare a set of monitoring sites for the development of a LUR model. 21 different sites in and around London were carefully selected in this way to represent either urban background locations, roadside locations, or regional background locations (See Figure 8).

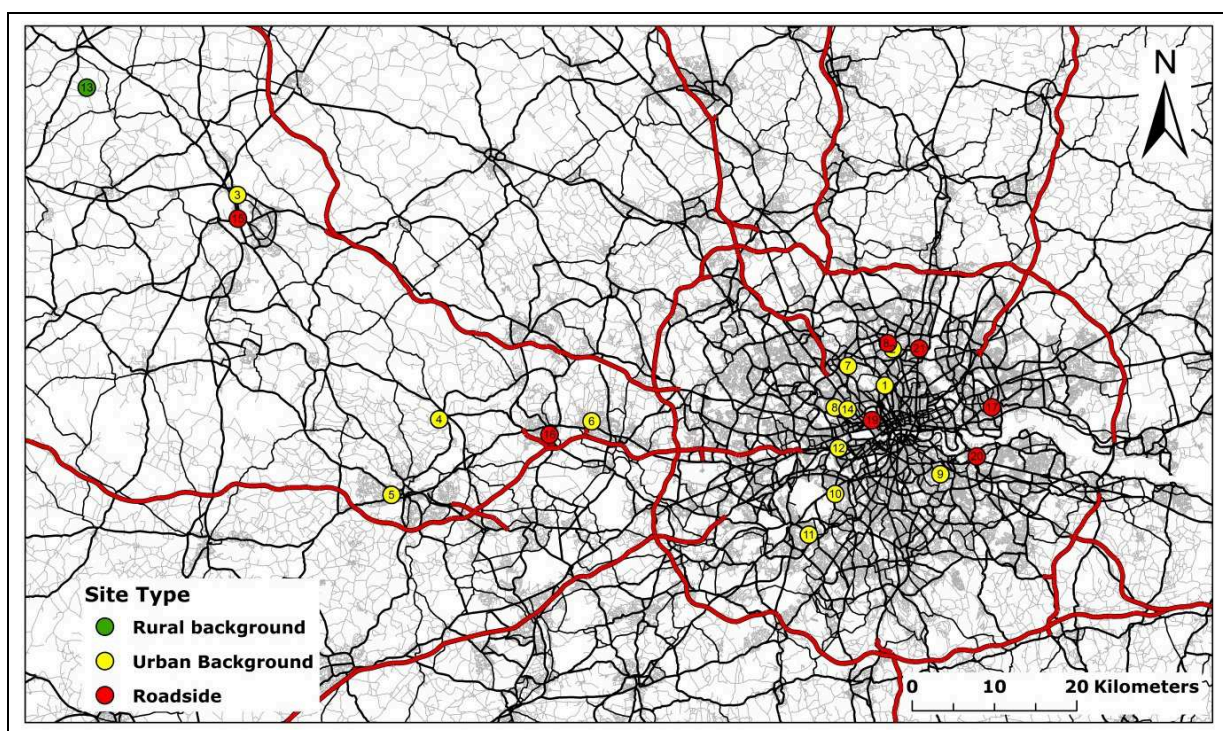


Figure 8: Monitoring sites selected for the ambient (ESCAPE study) campaign in London and along the Thames Valley. Road data source: Meridian.

Great care was taken to eliminate unwanted influences and potential confounders at the monitoring sites, for example from parking lots, fuel stations, overhanging trees or roofs, bars and restaurants, as well as bus stops. Locations were restricted to those, where the monitor could be installed at a minimum height of 1.5m and maximum height of 5m in order to produce comparable results. The 21 ESCAPE sites comprised of 7 traffic (or 'roadside') sites, 1 regional background site, and 12 urban background sites. One additional background site was functioning as a reference site and was – unlike the others – monitored all year round in 2 week intervals.

The first step for site selection was to identify possible monitoring sites and contact owners. Next, the candidate sites were visited and information was recorded on: distance to nearest road, distance to major road, vehicles passing per minute while on site, and height of building. Additionally pictures were taken of all four directions and the surrounding area was reviewed on google maps. In accordance to a protocol which outlined selection criteria, decisions were made on which sites to include. The selection was passed on for a second opinion to partners of the ESCAPE project (Brunekreef 2008). Abbreviated site descriptions and a representative picture of each of the final 21 sites are added in Appendix D.

All monitoring is undertaken with Harvard Impactor monitors, a commonly used PM mass detection device. At each site $PM_{2.5}$, as well as PM_{10} samples were collected, a $PM_{2.5}$ duplicate and a blank were additionally added. These four Harvard Impactors were connected to a pump unit. Each of the 20 sites (not the reference site) was monitored once in every season (winter, summer, intermediate) of year 2010 for a two-week period. During every seasonal campaign a pump unit with the attached PM monitors was installed at each site and removed again after two weeks. The run time was set to record 15 minutes every 2 hours in order to prevent filter overload. Air flow was tested and recorded at each Harvard Impactor at deployment and collection. Only five pump units were available for the 20 sites; a rotation scheme was therefore applied, so that four rounds with five simultaneously running monitors were undertaken in each of the three seasons. The reference site was visited every two weeks throughout the whole year. At each visit the filters were collected and replaced. Data from the reference site was utilised to calculate seasonal adjustments for all other sites by calculating an adjustment ratio of each seasonal measurement to the annual mean. These adjustments were then applied to seasonally weigh the three samples collected at each location to an annual average of year 2010 (Beelen & Hoek 2010).

All used filters were sent to a laboratory at IRAS (Institute for Risk Assessment Sciences) in Utrecht, the Netherlands for analyses. The $PM_{2.5}$ and PM_{10} concentrations measured at the different sites were subsequently used to calculate a LUR model for London and the Thames Valley area, which is described in chapter 4. Further details of the ambient modelling campaign are described in Eeftens et al. (2012), for which the doctoral candidate is co-author.

3.2.2. Description of Results

After analyses of the filter samples by colleagues from the ESCAPE study, the concentrations were provided to the doctoral candidate as adjusted annual concentrations. Annual average concentrations from the ambient monitoring campaign are presented in Figure 9. It can be observed

from the presented box plots that both for $PM_{2.5}$ and PM_{10} , the median concentrations and range are higher for traffic locations in comparison to background locations. For $PM_{2.5}$, mean concentrations of all sites are $11.21\mu\text{g}/\text{m}^3$, the SD is $2.71\mu\text{g}/\text{m}^3$ and the range is $14.18\mu\text{g}/\text{m}^3$. Results at a central London background site, Kensington and Chelsea - North Kensington (data source: LAQN: <http://www.londonair.org.uk/london>, accessed: 17th Oct. 2013) show in comparison a higher mean of $14.02\mu\text{g}/\text{m}^3$ (2010 average).

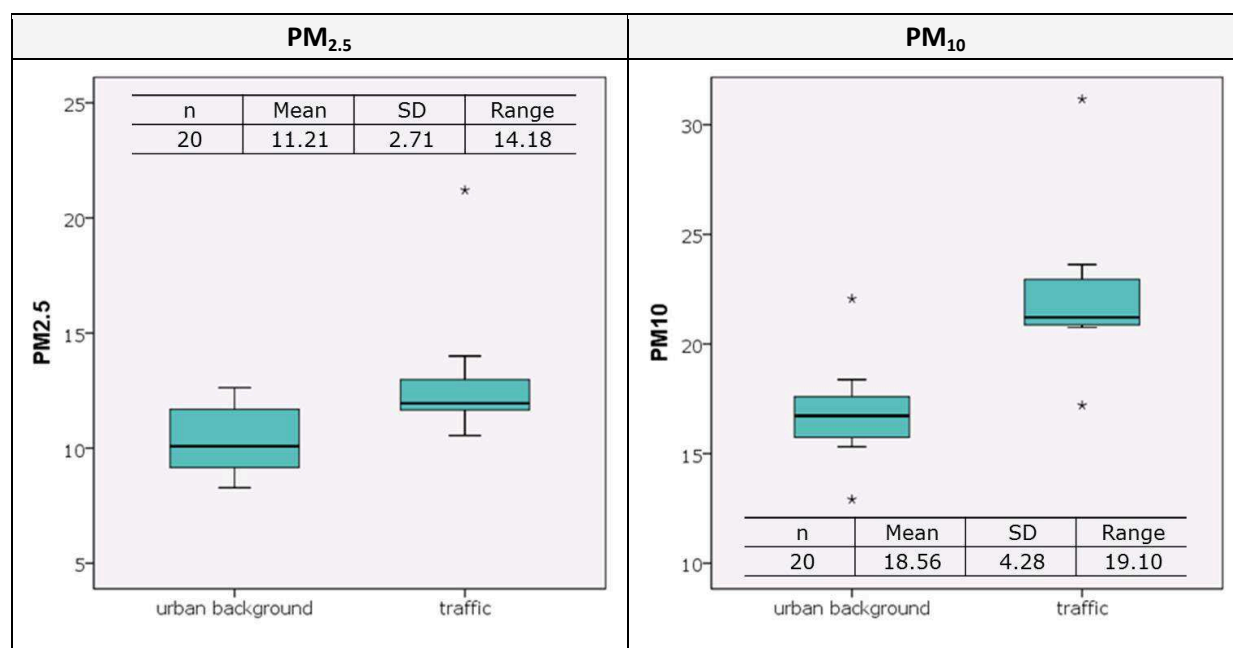


Figure 9: Annual concentrations for $PM_{2.5}$ and PM_{10} (both in $\mu\text{g}/\text{m}^3$) at background and traffic locations, collected during the ambient monitoring campaign. Basic statistics are added for both locations combined.

For PM_{10} , data collected during the ambient monitoring campaign show a mean concentration $18.56\mu\text{g}/\text{m}^3$, a SD of $4.28\mu\text{g}/\text{m}^3$ and a range of $19.10\mu\text{g}/\text{m}^3$. Results from the North Kensington monitoring station in comparison show a slightly higher mean of 20.36 for 2010.

Further monitoring results from the ESCAPE study with comparisons between different study centres are presented in Eeftens et al. (2012), for which the doctoral candidate is co-author.

3.3. Residential Indoor and Outdoor Monitoring Campaign

3.3.1. Monitor Set Up at Home Locations

Indoor-outdoor monitoring for home locations was undertaken in four different locations in London (see Figure 10). All homes are located in background locations, along minor roads, with the nearest major roads at least 50m away and behind closed building fronts. None of the four homes was influenced by environmental tobacco smoke (ETS). The indoor monitor was located in all homes at the main floor (where most activities take place during the day). It was taken care that indoor and outdoor monitors were positioned in approximately mirrored locations inside and outside. In all cases the outdoor monitor was located at the back of the house for security reasons. The outdoor monitors were located at an open air position (e.g. not under trees). It was furthermore taken care to avoid potentially confounding conditions, for example close to exhausts or open sand pits. Monitors were located at a height of at least 1m to avoid direct influence from re-suspended dust and soil from the ground.

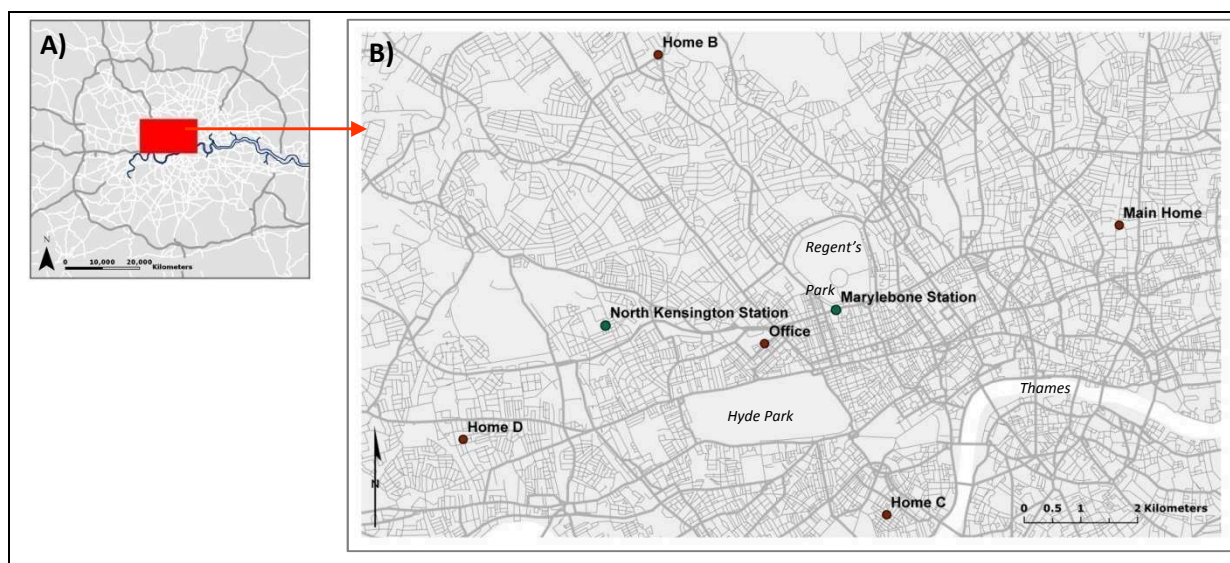


Figure 10: A) Map of Greater London, B) Map Extract of the Study Area for residential indoor and outdoor, as well as personal monitoring. Road data source: Meridian.

The home indoor monitor also functions as a proxy for personal exposure for this study's personal exposure monitoring campaign (for times at home indoors). This meant however that movement between rooms by the individuals at home could not be followed. Reasons were that all homes were relatively small, the monitors too big to carry around constantly, and the repeated movement and handling of the monitors could have changed particle concentrations. All times of home occupancy

were accompanied by a diary on a minute basis (recording start and end times). Individuals recorded numbers of occupants at the main floor level, as well as the following activities:

- Cleaning: including vacuum cleaning, dusting, wet cleaning, and major tidying activities (i.e. changing sheets) on the main floor level
- Cooking: any type that involves preparation of cooked food on the main floor level
- Sleeping: including naps and reading in bed etc. on any floor
- Special activities: not including sitting, standing or walking, e.g. DIY (no special activities however occurred)

Additionally, times of ventilation (number of open windows) were recorded. However, ventilation was changed rarely or only for a few minutes at any time, data was therefore not sufficient to be included in the following data description.

A description of the four home locations is added in Appendix E.

3.3.2. Residential Outdoor Concentration

Residential outdoor concentrations have been collected at all four study homes. Summary statistics are first presented for each of the homes, then by hour of day and for seasonal differences (home A only).

PM concentrations at the different home outdoor locations are defined by spatial influences, for example the distance to the next major road, as well as by temporal influences, such as differences in background concentrations over time, or weather related temporal changes. The main purpose for collecting this data was to provide outdoor PM concentrations as a predictor for indoor pollution, where for most people much of the daily exposure takes place.

Summary statistics of $PM_{2.5}$ and PM_{10} for home outdoor locations are presented in Table 13. For $PM_{2.5}$ all median values at the different homes lie within a small range of between $4.68\mu\text{g}/\text{m}^3$ and $5.86\mu\text{g}/\text{m}^3$. The IQR shows only moderate differences between the locations (between $3.42\mu\text{g}/\text{m}^3$ and $6.43\mu\text{g}/\text{m}^3$). The 99th percentile remains with values between $12.43\mu\text{g}/\text{m}^3$ and $16.21\mu\text{g}/\text{m}^3$ as well moderate for all four locations.

	PM _{2.5}				PM ₁₀			
	Home A	Home B	Home C	Home D	Home A	Home B	Home C	Home D
n of Days	39	11	7	8	39	11	7	8
Median (Mean)	5.55 (6.15)	5.28 (5.83)	5.86 (6.92)	4.68 (5.64)	16.33 (17.75)	16.10 (16.49)	17.30 (18.70)	15.10 (17.44)
IQR (SD)	4.51 (3.60)	5.10 (3.27)	6.43 (3.77)	3.42 (8.63)	12.6 (19.32)	11.7 (8.05)	11.9 (11.12)	9.1 (34.03)
99th P. (Range)	15.58 (116.23)	12.85 (59.97)	16.21 (23.05)	12.43 (352.80)	40.40 (2280.20)	32.60 (222.70)	40.20 (451.10)	34.20 (1898.80)

Table 13: Outdoor concentrations of PM_{2.5} and PM₁₀ (both in µg/m³) for homes A, B, C, and D, based on minute data.

For PM₁₀ (as for PM_{2.5}) little difference can be observed between the homes. Median values lie between 15.10µg/m³ and 17.30µg/m³. IQR lie between 9.1µg/m³ and 12.6µg/m³, the 99th percentiles, lie a little wider apart, between 32.6µg/m³ and 40.4µg/m³.

In summary it can be said that outdoor median concentrations for both particle size fractions and at all four homes are relatively low, and variability is moderate. Additionally, little differences could be observed between the study homes, despite different time periods of monitoring. Low concentrations and low variability probably reflect the fact that all four locations are located in a background area, away from direct source influences.

Differences between Hours of Day

It can be seen in Figure 11 that all locations show a rise in median values during the early hours of the day, resulting in a peak between about 7am and 9am. This communality indicates a general trend at all locations, despite temporal and spatial differences. All homes show a decrease in values during the late morning and mid-day. PM concentrations in the evening show different trends at the different home locations and for different size fractions, most median concentrations show however higher concentrations in the late afternoon and evening than during the early afternoon. In some cases an evening peak can be observed.

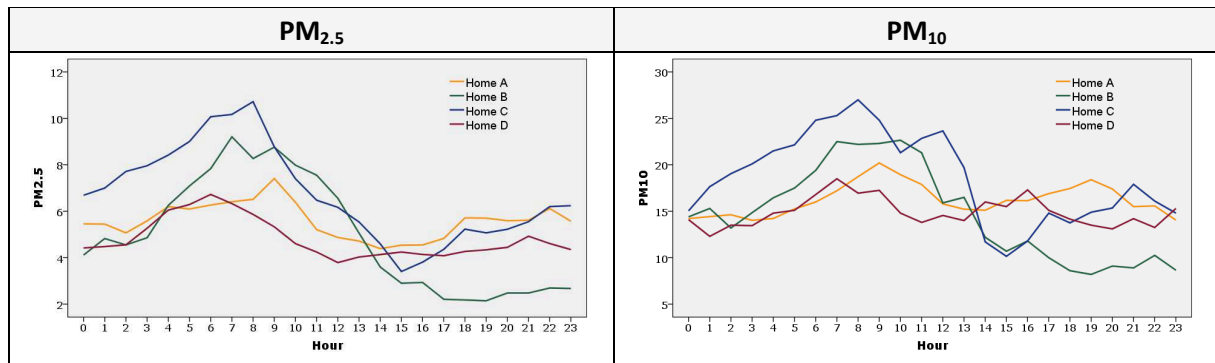


Figure 11: Median (minute) outdoor concentrations at the four study homes per hour of the day for $PM_{2.5}$, and PM_{10} (both in $\mu g/m^3$).

The variability of outdoor concentrations during the course of a day has been analysed by other studies. One study by Hoek et al. (2008) looked at particle numbers at 152 homes in four European cities. The study showed outdoor peak values during morning rush hour times, similar to results in this thesis. Hoek et al. (2008) however also observed another, less pronounced peak during evening rush hours. An evening peak for data from this thesis could for example be observed as well for PM_{10} concentrations at home D. Only small datasets were collected for homes B, C, and D, the presented results by hour of day may therefore not be fully representative for the locations.

Seasonal Differences

One of the main reasons to monitor indoor and outdoor concentrations at one of the homes over the course of almost a year was to examine potential influence of seasonality. At home A monitoring data for 8 days in spring, 16 days in summer, 8 days in autumn, and 7 days in winter were collected. The seasons were defined following the monthly definition from the UK met office: spring = March to May, summer = June to August, autumn = September to November, winter = December to February. It is understood that some days may not represent the typical weather pattern of the season; the definition is however widely used and accepted.

In Table 14, $PM_{2.5}$ and PM_{10} concentrations are presented per season. Differences between median levels between the seasons are small and the highest median was observed in winter with $7.28\mu g/m^3$, followed by the intermediate seasons. The lowest median concentrations has been recorded for the summer median concentrations with $4.4\mu g/m^3$. The IQR is highest in autumn ($8.37\mu g/m^3$), and second highest in winter ($4.85\mu g/m^3$), summer shows the lowest IQR ($3.03\mu g/m^3$), spring the second lowest IQR ($3.45\mu g/m^3$).

	PM _{2.5}				PM ₁₀			
	Spring	Summer	Autumn	Winter	Spring	Summer	Autumn	Winter
Days	8	16	8	7	8	16	8	7
Median (Mean)	6.39 (6.66)	4.40 (4.84)	5.63 (6.56)	7.28 (7.11)	17.95 (20.16)	15.93 (17.95)	16.98 (16.86)	15.00 (15.56)
IQR (SD)	3.45 (2.44)	3.03 (2.50)	8.37 (5.80)	4.85 (3.14)	14.0 (9.14)	9.9 (26.67)	24.6 (23.57)	11.3 (7.68)
99 th P. (Range)	13.47 (17.09)	12.52 (37.20)	27.46 (116.23)	15.64 (16.12)	41.58 (76.40)	37.82 (2276.60)	48.34 (1342.20)	34.53 (146.40)

Table 14: PM_{2.5} and PM₁₀ concentrations collected with the outdoor monitor at home A per season, based on minute data.

Concentrations of PM₁₀ per season show similar median concentrations for the different seasons, which only vary between 15µg/m³ and 17.95µg/m³. Highest median concentrations were in this case recorded during spring (17.95µg/m³), followed by autumn (16.86µg/m³), then summer (15.93µg/m³). The lowest concentrations have been observed during winter (15µg/m³). The IQR is however again (as for PM_{2.5}) highest in autumn and lowest in summer.

Several other studies have looked into seasonal differences for PM in ambient air. They generally show higher outdoor concentrations in winter than in summer, as for example described for Ipswich and Norwich (England) as part of the ECRHS II (European Community Respiratory Health Survey II) study (Arx et al. 2004). A study by Nawrot et al. (2007) presents data for temperature-defined seasons in Belgium, and finds again higher average PM₁₀ concentrations in winter, than in summer. Results for the intermediate seasons are harder to interpret or compare, as weather patterns are less distinctive. For PM_{2.5}, results in this study directly agree with findings in the literature with higher median concentrations in winter compared to summer. PM₁₀ concentrations show however a slightly higher median concentration for summer compared to winter. The number of days sampled per season is however small and might not have been sufficient to capture a representative range of concentrations.

3.3.3. Residential Indoor Concentrations

In the following paragraphs PM_{2.5} and PM₁₀ concentrations recorded inside the four homes are described and compared. In the following section first basic statistics are presented for each of the four study homes; then, results are presented per hour of day. In addition, concentrations are investigated for different occupancy levels, as well as times of cooking and cleaning.

Results for indoor concentrations of PM_{2.5} and PM₁₀ per home are presented in Table 15. For PM_{2.5}, homes A, B, and D show generally low, as well as similar concentrations for the median (between 4.16µg/m³ and 4.33µg/m³) and for the IQR (between 3.00µg/m³ and 3.56µg/m³). Their 99th

percentiles lie between 20.25 $\mu\text{g}/\text{m}^3$ and 50.52 $\mu\text{g}/\text{m}^3$. Home C however shows almost double the median (7.86 $\mu\text{g}/\text{m}^3$) and IQR (6.99 $\mu\text{g}/\text{m}^3$) concentrations. The 99th percentile lies with 316.61 $\mu\text{g}/\text{m}^3$ as well much higher than what observations at the three other homes show.

	PM _{2.5}				PM ₁₀			
	Home A	Home B	Home C	Home D	Home A	Home B	Home C	Home D
n of Days	39	11	7	8	39	11	7	8
Median (Mean)	4.28 (6.20)	4.16 (4.55)	7.86 (17.22)	4.33 (6.25)	10.18 (21.21)	8.8 (10.63)	23.7 (60.82)	9.4 (21.27)
IQR (SD)	3.56 (19.00)	3.14 (3.48)	6.99 (50.98)	3.00 (9.16)	10.4 (132.72)	6.2 (8.43)	36.5 (159.46)	8.8 (57.27)
99th P. (Range)	34.85 (652.00)	20.25 (44.68)	316.61 (564.73)	50.52 (175.41)	173.11 (6526.40)	43.52 (112.7)	808.15 (2659.20)	298.01 (1061.90)

Table 15: Indoor PM_{2.5} and PM₁₀ (both in $\mu\text{g}/\text{m}^3$) concentrations at homes A, B, C, and D, based on minute data.

For PM₁₀ (as for PM_{2.5}) median and IQR concentrations for homes A, B, and D are low, as well as similar. The median concentrations for these three homes lie between 8.8 $\mu\text{g}/\text{m}^3$ and 10.18 $\mu\text{g}/\text{m}^3$, of which the lowest median was detected in home B and the highest home A. The IQR at these three homes lie between 6.2 $\mu\text{g}/\text{m}^3$ at home B and 10.4 $\mu\text{g}/\text{m}^3$ at home A. Home C however shows again much higher concentrations both for the median (23.7 $\mu\text{g}/\text{m}^3$), as well as for the IQR (36.5 $\mu\text{g}/\text{m}^3$). The 99th percentile is with 808.15 $\mu\text{g}/\text{m}^3$ for home C is also highest; the second highest 99th percentile was observed at home D with 298.01 $\mu\text{g}/\text{m}^3$; the third highest 99th percentile at home A with 173.11 $\mu\text{g}/\text{m}^3$, and the lowest 99th percentile was observed at home B.

The particularly high median concentrations and variability at home C could have many potential causes. Outdoor PM concentrations at home C were low and had little variability (see above); it is therefore much more likely that the cause for these high and highly variable concentrations originates inside the home, as for example from indoor source activities (such as people's movement or cooking).

Temporal Patterns

In Figure 12, the indoor median concentrations for all four study homes are presented per hour of day. All homes show a morning peak for all size fractions after lower night-time values and before declining values during late morning and early afternoon. The exact timing of the peak however varies between 7am and 9am, the intensity of the peak also varies between homes and is generally least pronounced at home D and most pronounced at home C. Median concentrations in the evening are higher than in the afternoon and for some homes a clear evening peak can be observed,

as for example at home D. Evening peaks for the other homes are however only clearly visible for PM_{2.5}.

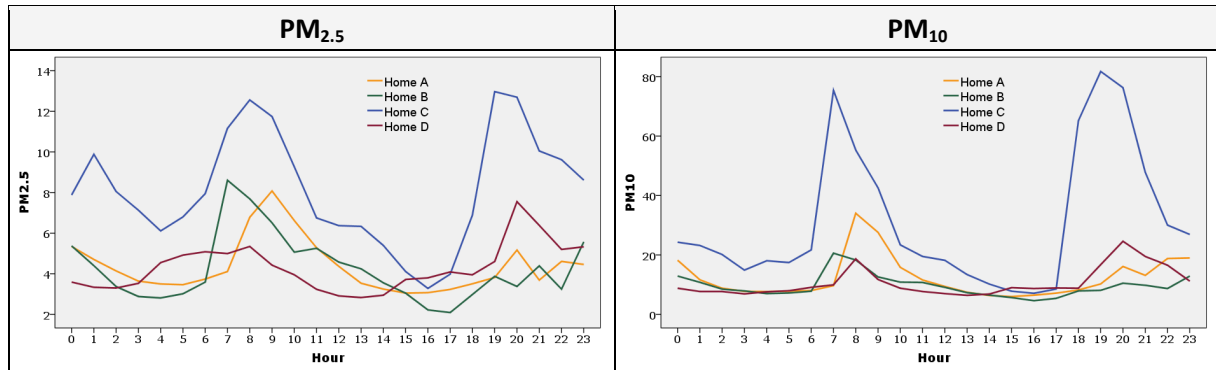


Figure 12: Median (minute) indoor concentrations at the four study homes per hour of the day for PM_{2.5}, and PM₁₀ (both in µg/m³).

The patterns of the indoor median values during the course of a day are very similar to the patterns described for outdoor PM throughout the day (see above). For outdoor concentrations, a connection to rush-hour traffic has been suggested (Hoek, Kos, et al. 2008). For indoor concentrations, increased concentrations are likely to be connected to increased human activity following a working day pattern (high activity before leaving the house in the morning and after coming back from work). It should again be noted, that homes B, C, and D have been recorded during few days and over short time period, which may have influenced the results.

3.3.4. Influences of Home Occupancy on Indoor Concentrations

Occupancy has been shown to increase indoor PM concentrations in several studies and times of no occupancy, as well as times of sleep are commonly associated with lower levels of indoor PM (Hänninen et al. 2004; Kamens et al. 1991). Several categories of occupancy were recorded in the diary during indoor monitoring. The categories are:

- a) No occupancy,
- b) Sleep (all people in the house are sleeping),
- c) 1 person on the main floor,
- d) 2 people on the main floor,
- e) 3 or more people on the main floor (maximum recorded: 5 people).

Summary statistics for indoor air pollution per category of occupancy are presented in Table 16. The presented data is pooled for all four home locations. Combining the datasets meant a bigger dataset with more occupancy differences for comparison. On the other hand it is possible that some differences between the categories might be due to other differences between the homes (not related to occupancy).

	PM _{2.5}					PM ₁₀				
	No occ.	Sleep	1 Person	2 People	3+ People	No occ.	Sleep	1 Person	2 People	3+ People
n of Minutes	20613	25251	10567	3021	1541	20613	25251	10567	3021	1541
Median (Mean)	4.03 (5.05)	4.25 (5.13)	4.94 (9.40)	7.28 (25.01)	9.89 (31.71)	8.50 (12.08)	9.60 (13.20)	16.05 (38.16)	32.20 (111.61)	73.70 (155.95)
IQR (SD)	3.50 (3.99)	3.17 (4.60)	3.87 (27.43)	9.89 (66.34)	8.57 (93.62)	7.3 (11.94)	8.0 (13.18)	20.2 (106.28)	53.7 (469.14)	81.90 (310.55)
99th P. (Range)	23.65 (44.45)	22.94 (108.09)	91.80 (570.73)	320.84 (651.25)	528.53 (564.40)	63.47 (379.18)	69.59 (175.50)	433.80 (2712.00)	814.00 (6522.45)	1830.35 (2658.30)

Table 16: Indoor Concentrations per level of occupancy for PM_{2.5} and PM₁₀ (both in µg/m³) in minutes. No occ. = No occupancy

For PM_{2.5}, median concentrations are the lowest at times for no occupancy (4.03µg/m³) and the second lowest, by a small margin, for times of sleep (4.25µg/m³), median concentrations then rise successively with the number of people on the main floor from one (4.95µg/m³) over two (7.28µg/m³) to three or more (9.89µg/m³). The IQR is slightly lower during sleeping times (3.17µg/m³) than during no occupancy (3.50µg/m³), IQR during occupancy by one person is again a bit higher (3.87µg/m³), the highest IQR was recorded when two people were present (9.89µg/m³) and the second highest when three or more people were on the main floor (8.57µg/m³). The 99th percentile is the lowest for times of sleep with 22.94µg/m³ and for no occupancy with 23.65µg/m³. Concentrations for the 99th percentile then rise steeply with the number of people. A 99th percentile of 91.80µg/m³ was recorded while one person was around, 320.84µg/m³ were recorded while two people were on the main floor, and the highest 99th percentile was recorded when three or more people were around was highest with 528.53µg/m³.

Summary statistics for PM₁₀ (as for PM_{2.5}) show that median concentrations are lowest for no occupancy (8.50µg/m³) and sleeping times (9.60µg/m³); then they successively rise by number of people in the main floor (1 person: 16.05µg/m³, 2 people: 32.20µg/m³, 3+ people: 73.70µg/m³). The IQR shows the same succession of concentrations from no occupancy (7.3µg/m³), over sleeping times (8.0µg/m³), one person (20.2µg/m³), two people (53.7µg/m³), to three or more people (81.90µg/m³). The same succession is again displayed by the 99th percentile, showing the lowest values for no occupancy (63.47µg/m³) and sleeping times (69.59µg/m³). For one person the 99th

percentile is $433.80\mu\text{g}/\text{m}^3$, for two people it is $814.00\mu\text{g}/\text{m}^3$ and for three or more people it is $1830.35\mu\text{g}/\text{m}^3$.

In summary, the median concentrations, as well as the variability of the dataset are lowest during times where people are not at home or sleeping. Median concentrations and variability generally rise with the number of people present on the main floor level. These observations mirror what has been found by other studies. Hänninen et al. explored the influence of different indoor characteristics on indoor $\text{PM}_{2.5}$ concentrations in a regression analysis as part of the EXPOLIS-study (Air Pollution Exposure in European Cities). The study found that the number of family members significantly influenced concentrations of $\text{PM}_{2.5}$ (Hänninen et al. 2004). At home C most of the time recorded as active occupancy was occupied by four people, compared to one or two at the other homes. The higher concentrations generally correlated with higher number of people could therefore be the explanation of high indoor concentrations at home C. It should be noted that for the comparison of occupancy levels in this section other sources were not excluded for the relevant time periods and may have influenced results.

3.3.5. Influences of Cooking and Cleaning on Indoor Concentrations

Cooking

Several studies have been conducted that look at PM concentrations during cooking activities (see chapter 2). These studies show much higher particle concentrations during cooking activities than have been found for average home indoor concentrations. Additionally it has been observed that the recovery time after cooking to normal levels can take hours. Cooking is therefore a major indoor source and can significantly change home indoor PM exposure (Glytsos et al. 2010; He et al. 2004; Olson & Burke 2006). During indoor monitoring for this thesis, cooking activities took place in homes A, C, and D.

Cooking Times

Figure 13 shows box plots of $\text{PM}_{2.5}$ and PM_{10} concentrations for times of no cooking activity, times of cooking, and for the 60min after cooking had finished. Times of no cooking activity were for this comparison chosen only for times during which at least one person was at home and not sleeping (60min after cooking were as well excluded). Home A shows for both $\text{PM}_{2.5}$ and PM_{10} a clear increase of median concentrations from no cooking, over cooking, to hour after cooking. For home C, both $\text{PM}_{2.5}$ and PM_{10} show as well highest concentrations for the hour after cooking. For home D, median

concentrations for all three categories (not cooking, cooking, hour after cooking) are relatively similar. The variability (box and whisker lengths) of concentrations is however highest for times of cooking, followed by the hour after cooking. Lowest variability can be observed for times without cooking.

In summary, results about the effect of cooking and the hour immediately after are not fully conclusive. A more detailed presentation of results is therefore given in Figure 14. The figure shows all single cooking events (above 10min duration) for the different homes and both PM size fractions. The diagrams start one hour before the start of cooking activities and continue for up to four hours after the start of cooking.

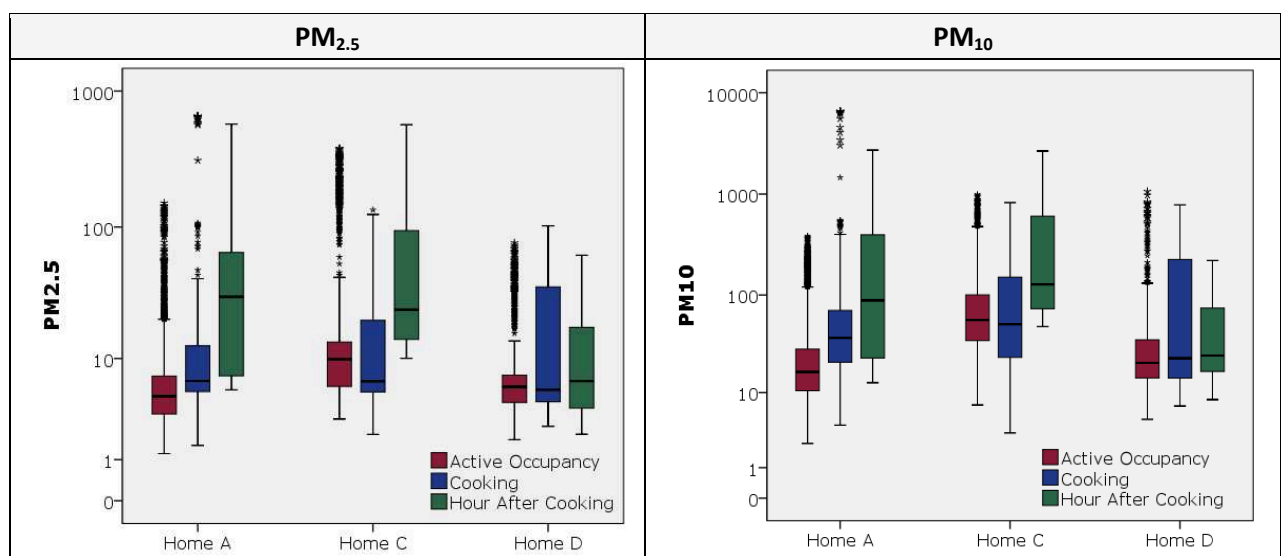


Figure 13: Indoor concentrations per home for times of 1) active occupancy (at home, not sleeping), 2) without cooking activity, 3) for times with cooking activity, and 4) for the hour after cooking; for $PM_{2.5}$, and PM_{10} (both in $\mu g/m^3$). Note: concentrations are presented at a log scale.

All cooking events show typical examples of cooking peaks, as described by He et al. (He et al. 2004), with a steep rise in concentrations when cooking activity starts and a slow return to background values afterwards (recovery time).

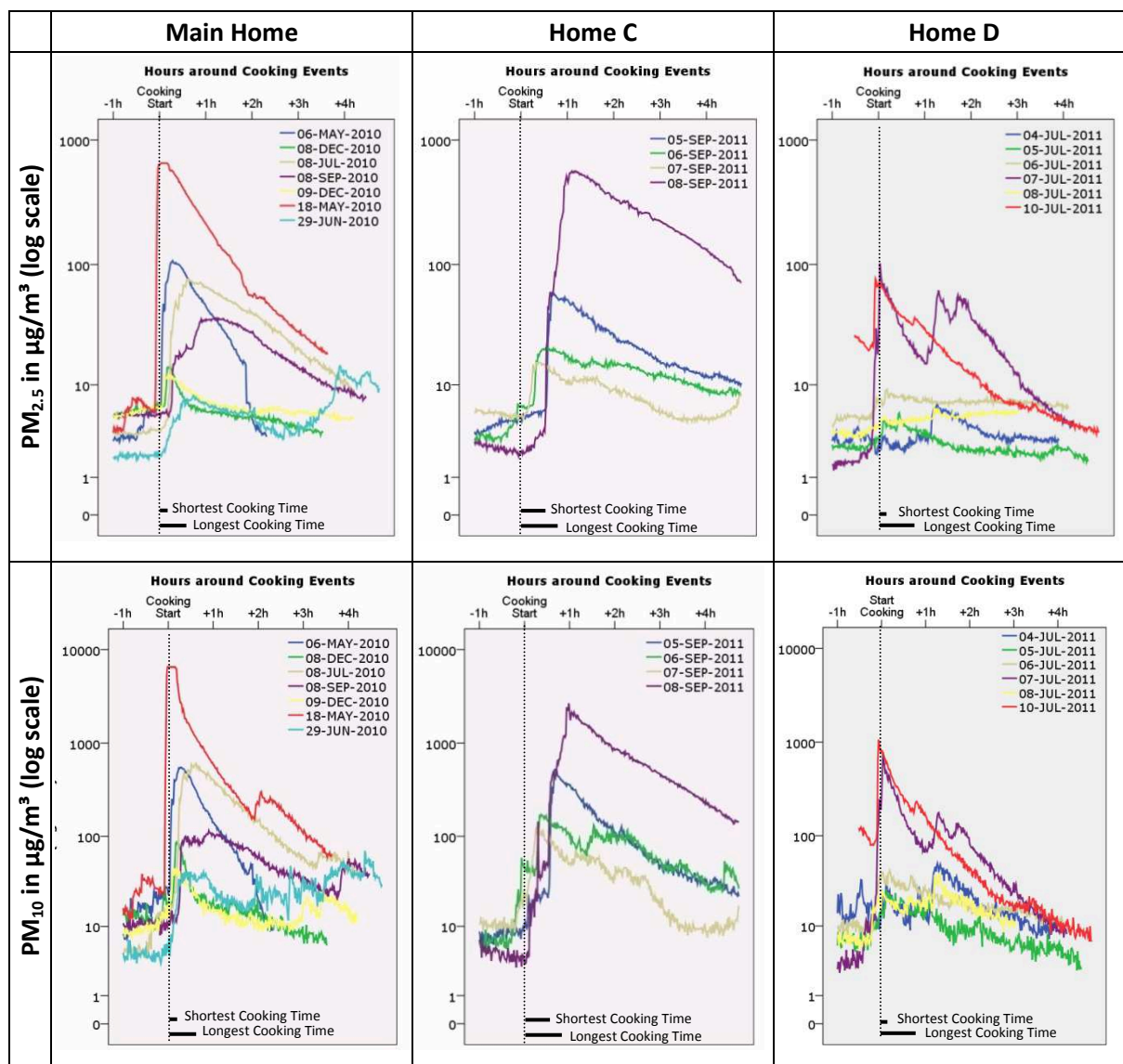


Figure 14: Home indoor concentrations of $PM_{2.5}$, and PM_{10} (both in $\mu g/m^3$) for homes A, C, and D for cooking events, starting from one hour previous to the start of cooking activity and up to four hours after the start of cooking. Minimum and maximum cooking times are added.

The presented cooking events for home A suggest that cooking related PM concentrations are picked up immediately by the monitor, which is located close to the kitchen. In the examples for home C the diary recorded cooking activity starts before the PM peak can be observed for the PM data in the graph. The indoor monitoring device in home C is located in a different room at the other end of the flat, which could have caused this lag time. Lag times for cooking related concentrations within a home have for example described by Abt et al. (2000). A lag time for cooking related concentrations at home C can explain why in Figure 13 median cooking related concentrations remain comparably low.

For home D, concentrations increase immediately after cooking started. The monitor is located in the same open-plan room as the kitchen, which allows for a very quick response to cooking related concentration changes. In fact for some days it seems the cooking peak was registered by the monitor a few minutes before cooking activity was noted down in the diary.

The differences in maximum concentration during cooking peaks has been described in several studies, as presented in the literature chapter 2 and are mainly caused by differences in type of cooking. The results presented here match well with results from other studies in range and variability (Buonanno et al. 2009; Buonanno et al. 2010; Glytsos et al. 2010; He et al. 2004; Olson & Burke 2006). In an experimental study focused on different cooking types by Olsen & Burke for example, median concentrations for PM_{2.5} were only 7.4µg/m³ for cooking in a microwave oven, compared to grilling with 155µg/m³ and burnt food with 231µg/m³.

Cleaning

Several studies found that cleaning increases PM concentrations inside residences (Abt, Catalano, et al. 2000; Kamens et al. 1991; Meng et al. 2009; Thatcher & Layton 1995). For this study cleaning events were only recorded for home A and only on five different days. Even though the data is therefore limited, results (see Table 17) clearly show increased concentrations during cleaning times, compared to other times of ‘active occupancy’ (at least one non-sleeping person on the main floor).

	PM _{2.5}		PM ₁₀	
	No Cleaning	Cleaning	No Cleaning	Cleaning
n of Minutes	13988	99	13988	99
Median (Mean)	5.70 (15.54)	12.17 (12.28)	21.30 (68.53)	50.60 (71.52)
IQR (SD)	6.06 (50.51)	9.23 (5.48)	36.80 (261.48)	20.6 (52.37)
(Range)	651.58	18.27	6525.40	195.80

Table 17: Indoor concentrations at home A for times of active occupancy (not sleeping) with and without cleaning activity for PM_{2.5} and PM₁₀ (both in µg/m³), based on minute concentrations.

Median concentrations for PM_{2.5} are increased during cleaning with 12.17µg/m³ compared to 5.70µg/m³ during non-cleaning times. The same pattern can be found for PM₁₀: median concentrations during cleaning are raised to 50.60µg/m³, compared to 21.30µg/m³ during non-cleaning times. Comparisons between the IQR is difficult, due to the different amount of data points between cleaning and times without cleaning (99 data points compared to 13,988 respectively).

3.4. Personal Monitoring Campaign

3.4.1. Introduction

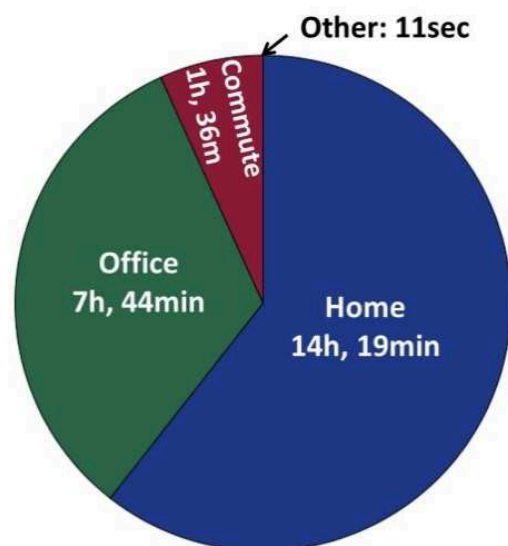


Figure 15: Average time spent in different MEs for individual A (only days with >75% of minutes in the day were included).

Personal exposure was altogether recorded by four different individuals: A, B, C, and D. All four individuals were between 25 years and 40 years of age, individuals A, B, and C are female, individual D is male. Individual A (the doctoral candidate) recorded personal exposure during 43 working days. The main microenvironments (MEs) visited during these days were the home ME, work ME (office), and times in commute. The average time spent in each of these MEs can be seen in Figure 15. On average 14 hours and 19 minutes were spent at home, 7 hours and 44 minutes in the office and 1 hour and 36 minutes in commute. Only on average 11 seconds were spent in other MEs (e.g. a friend's home, or a doctor's practice).

Results from the UK National Time Use Survey (Source: Office of National Statistics, Data Archive) shows in comparison that the average adult spends 69 minutes each day in transport, which is 27 minutes shorter than observed for individual A. Time spent working by fully employed adults is on average about 7 hours and 30 minutes, which is as well a little shorter compared to average times at work for individual A.

Individuals B, C, and D only recorded between two and four working days of personal monitoring each. During these days they spent between 1 hour 20 minutes and 1 hour 50 minutes per day in commute, and between 5 hours 35 minutes and 8 hours 55 minutes at work. Almost all remaining time was spent at home.

All four individuals clearly spent most time at home, followed by the time in the office. Time spent in commute in London is commonly longer than 1 hour (according to the UK time use survey 2000), which has been reflected by all four individuals, modes of commute however differ. For individual A, two different main commuter modes were used: bus and cycling. On 24 days the entire commute was undertaken by bicycle. On 19 days the main mode of commute was bus. The bus commute

comprises of two bus routes with walking to and from, as well as in between stops when changing buses. The commute of individual B was undertaken by one bus with walking to and from the bus stop. Individual C commuted to work by bus (plus walking to and from bus stops). On the return journey individual C walked home. Individual D commuted by bus and underground with walking to and from, as well as in between stops. During all personal monitoring, PM concentrations were recorded for 65 journeys on bus (counting each bus separately within one commute), for 50 cycling journeys, and for 74 periods of outdoor walking (mostly in conjunction with bus commutes).

The monitor was constantly carried or kept close during personal monitoring times when outside the home environment. During periods in the indoor home ME, concentrations were for practical reasons (noise of two running monitors, space limitations etc.) recorded by the indoor monitor (see indoor-outdoor monitoring campaign, above). Times spent in different MEs were recorded in a diary (by start and end minute). During journeys locations were recorded by a GPS.

Typical exposure variability during the day

In Figure 16 an example for daily exposure from individual A is presented (9th December 2010). Simultaneously monitored indoor and outdoor concentrations are added for comparison (Personal exposure equals indoor concentrations during times at home). Night times at home show low concentrations for the indoor/personal monitor. Concentrations rise when getting up in the morning, especially for PM₁₀. During the commute the individual experienced peak exposures. The commute was taken mostly by bus in several steps: walk-bus1-walk-bus2-walk. Some variability can be observed throughout the commute, which may be connected to changes in transport modes. Exposure in the office ME is low. For PM_{2.5} the concentration during office times are lower than at the unoccupied home location. For PM₁₀, concentrations in the office are similar to concentrations monitored at the unoccupied home for the same time. Cooking took place soon after the individual returned home, which can be observed as a small peak by the indoor/personal monitor.

For most types of MEs different locations were visited (different buses, different homes etc.) by different individuals. Only the office was shared between all individuals. Despite different locations, types of MEs however generally share some common characteristics (such as cooking source at home, influence of traffic during bus rides etc.). In Figure 17 concentrations recorded for personal exposure in different types of MEs are presented. It can be observed that the home ME shows for both PM_{2.5} and PM₁₀ the highest range for concentrations. Median concentrations at home remain however below concentrations recorded for all outdoor and transport related MEs.

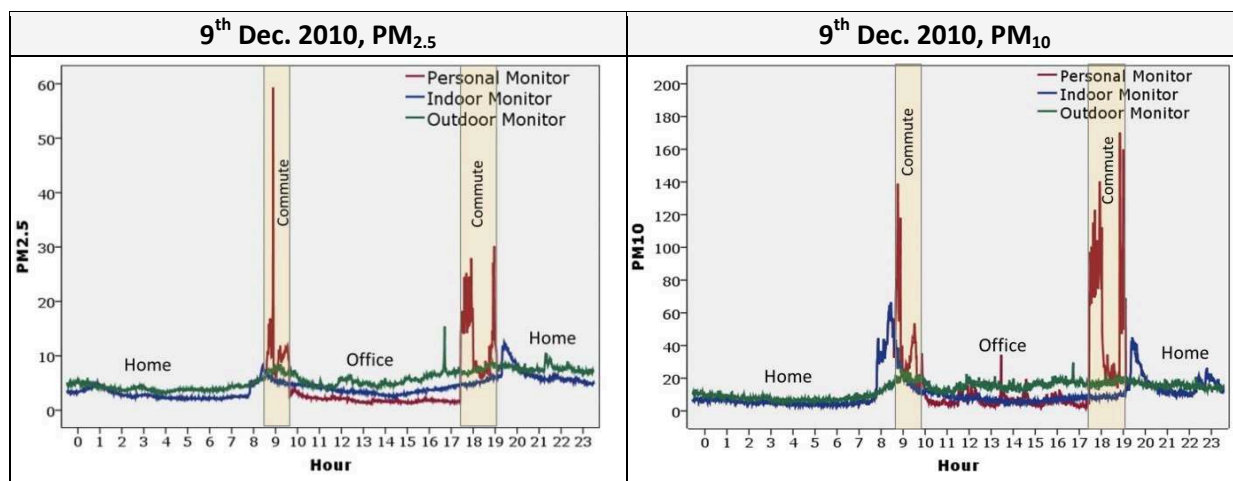


Figure 16: Example day of personal, indoor and outdoor monitoring (9th Dec. 2010) for $PM_{2.5}$ and PM_{10} using minute concentrations (both in $\mu g/m^3$) for individual/ home A.

Concentrations in the office show clearly the lowest median concentrations (both for $PM_{2.5}$ and PM_{10}). Concentrations for the remaining five MEs were mostly collected during commutes. The transport mode with the lowest concentration is walking. Walking exposure has as well been shown by McNabola et al. (2009) to be generally lower than direct in-traffic concentrations due to the horizontal displacement from the kerb. The highest median concentration was recorded for outdoor stationary locations, which mostly comprise of bus stops.

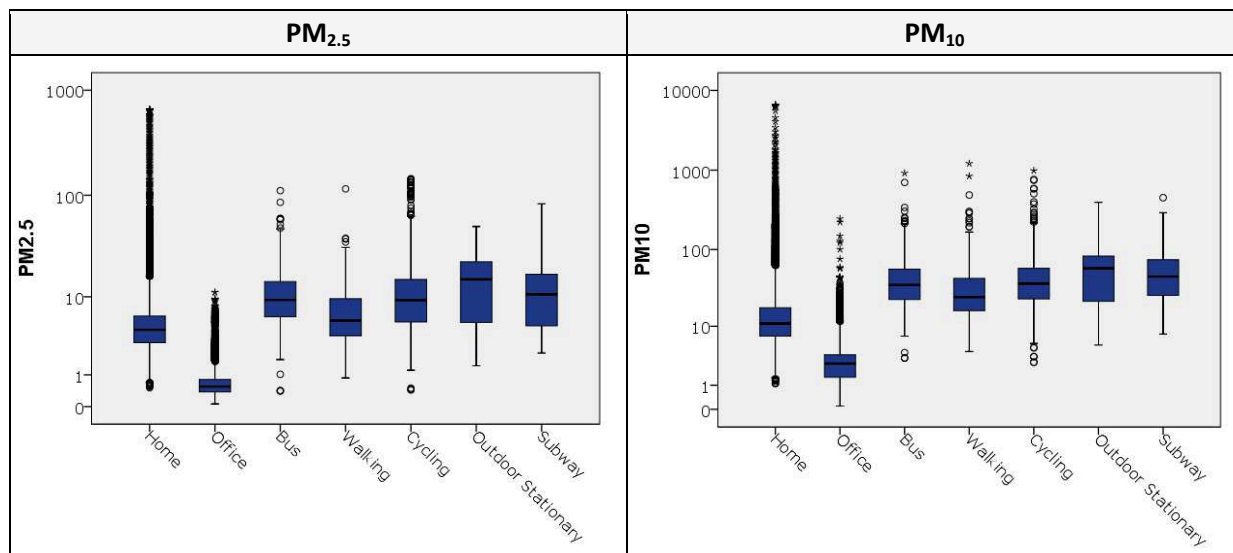


Figure 17: Minute concentrations by type of microenvironment for personal exposure of $PM_{2.5}$ and PM_{10} (both in $\mu g/m^3$).

Note: the Y axes are presented in log scale.

The comparably higher concentration at these locations could be related to high traffic sources along bus routes, as well as increased concentrations when buses idle at the stops. Median concentrations and variability for cycling, in-bus, as well as in the (above-ground) subway are similar for both particle size fractions and lie in between median results for walking and outdoor stationary locations.

Concentrations for each type of ME vary by location and type of situation. During personal exposure monitoring different locations have been visited for each type of ME. In the following sections all locations visited per each type of ME are presented. Locations have however been visited for different lengths of time and for different number of repeats and several locations have been visited only once or twice. For the following data description all locations are presented in the tables and maps. Only locations that were visited at least three times are however discussed in the text, as little information can be drawn from such small samples. Results of small samples are presented in the tables in grey.

3.4.2. Stationary Outdoor Locations

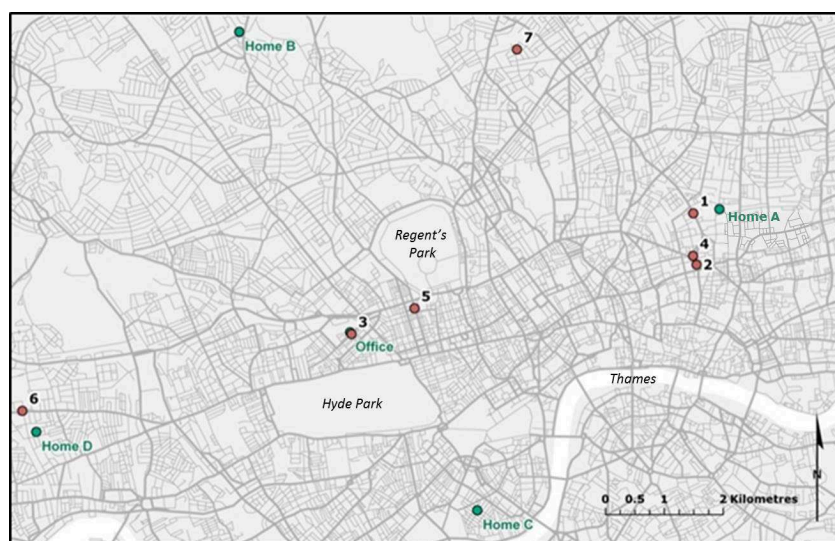


Figure 18: Central London, showing stationary outdoor locations visited during the personal exposure monitoring campaign, as well as homes A, B, C, and D, and office locations. Road data source: Meridian.

As part of the locations visited during personal exposure monitoring times by individuals A, B, C, and D, personal $PM_{2.5}$ and PM_{10} concentrations were recorded in 7 different stationary outdoor locations; their locations are shown in Figure 18. Generally, outdoor locations are influenced by background PM levels, as well as different levels of local PM, such as road traffic. Locations 1 to 6 are

bus stops, located along medium size or major roads, and are therefore be influenced to some extent by the amount of road traffic, as a local source. Location 7 is a playground, located in a park and more than 200m away from the next larger road. The location is therefore expected to be mostly influence by background air.

Summary statistics and daily average traffic (DAT), according to LAEI traffic count data for road segments, are presented in Table 18. The highest median values for PM_{2.5} and PM₁₀, of 22.46µg/m³ and 71.94µg/m³ respectively, were measured at the location 2. Location 2 also experiences by far the highest DAT of more than 45,000 vehicles per day. Traffic at all other bus stop locations (1, 3, 4, 5, and 6) is with between 5,000 and 16,000 DAT much lower. Median concentrations of those five other bus stops differ substantially and lie between 1.92µg/m³ and 14.42µg/m³ for PM_{2.5} and between 9.30µg/m³ and 57.85µg/m³ for PM₁₀. Differences between these medians do not follow the amount of DAT at the different sites. Frequent stops of buses at these bus stops may at some locations have increased concentrations, despite comparably low traffic counts.

Outdoor Location	n	PM _{2.5}		PM ₁₀		DAT
	Days (Minutes)	Median (Mean)	IQR (SD)	Median (Mean)	IQR (SD)	All vehicles
1	8 (30)	11.60 (15.80)	21.54 (11.88)	50.80 (56.25)	50.15 (69.04)	15,692
2	11 (64)	22.46 (23.25)	13.22 (8.88)	71.94 (78.86)	38.00 (35.31)	45,846
3	11 (75)	14.42 (15.10)	7.59 (8.36)	57.85 (63.12)	48.78 (31.89)	5,013
4	5 (20)	7.51 (9.78)	8.51 (4.92)	24.95 (33.94)	39.94 (20.65)	5,767
5	2 (8)	1.92 (2.17)	1.24 (0.66)	9.30 (9.28)	1.85 (1.43)	12,722
6	4 (17)	2.97 (3.76)	3.45 (2.45)	16.30 (16.21)	14.45 (9.33)	15,157
7	1 (20)	3.23 (3.28)	0.41 (0.24)	18.60 (18.13)	4.55 (2.76)	-

Table 18: PM_{2.5} and PM₁₀ concentrations (in µg/m³ and per minutes) at stationary outdoor locations visited as part of the personal monitoring campaign, as well as daily average traffic (DAT). Traffic data source: LAEI.

There could be several reasons for the differences between concentrations and the lack of a correlation between concentrations and amount of traffic close to the site: 1) the average DAT might not be representative for the traffic at the time of monitoring (i.e. during rush-hour traffic). 2) the background levels may have been different during times of monitoring, 3) the amount of recorded data might not have been enough for a representative range of concentrations at the site. Further factors include for example meteorological influences.

3.4.3. Indoor Locations

The dataset of personal monitoring at home indoors is shared with the indoor-outdoor monitoring campaign. The results of times of occupancy for the four study homes and individuals (A, B, C, and D)

is equivalent to the personal monitoring spent at home and several aspects (i.e. times of sleep, effects of cooking etc.) have already been described as part of the chapter on indoor monitoring (page 94ff).



Figure 19: Central London, showing locations of indoor monitoring during the personal exposure monitoring campaign. Road data source: Meridian.

Summary statistics for times at which indoor concentrations form part of the personal monitoring campaign (summarised as times at home by individual including sleeping) are included in Table 19, together with summary statistics for other visited indoor locations.

All four individuals worked in the same office for which concentrations were recorded during 48 working days in total (mostly collected by individual A). The building is located at a busy road in Paddington, London (see Figure 19). All office rooms visited for this study are sealed and centrally air conditioned. Air enters into the system from the roof above the 4th floor. The monitor was located at the back of the desk of the monitoring individual. The individuals worked in three different rooms of the building, one on the first floor and two on the fifth floor. In all cases the location was away from communication areas or passages, the office environment is modern (few “dust traps”) and regularly cleaned. All offices were multi-occupant offices with between four and seven occupied desks.

Figure 20 shows box plots of minute concentrations recorded for different rooms in the office. Office room 1 was only occupied by individual A (Apr. – Nov. 2010), office room 2 was occupied by individual A (Dec. 2010 – Feb. 2011), as well as by individual B, and C. Office room 3 was only occupied by individual D. All three offices show similarly low concentrations, both for PM_{2.5} and for PM₁₀. Office 3 shows less variability compared to the other two rooms, which is related to the fact

that data for office room 3 was only collected during four days. Office 2 shows higher variability compared to office 1. Much of the variability can however be related to one day in February 2011 (28th). During that day parcels were packed for several hours in the office room, which may have raised concentrations due to resuspension. In summary, the centralised air conditioning unit appears to have distributed air with similarly low particle concentrations into the different office rooms at all times.

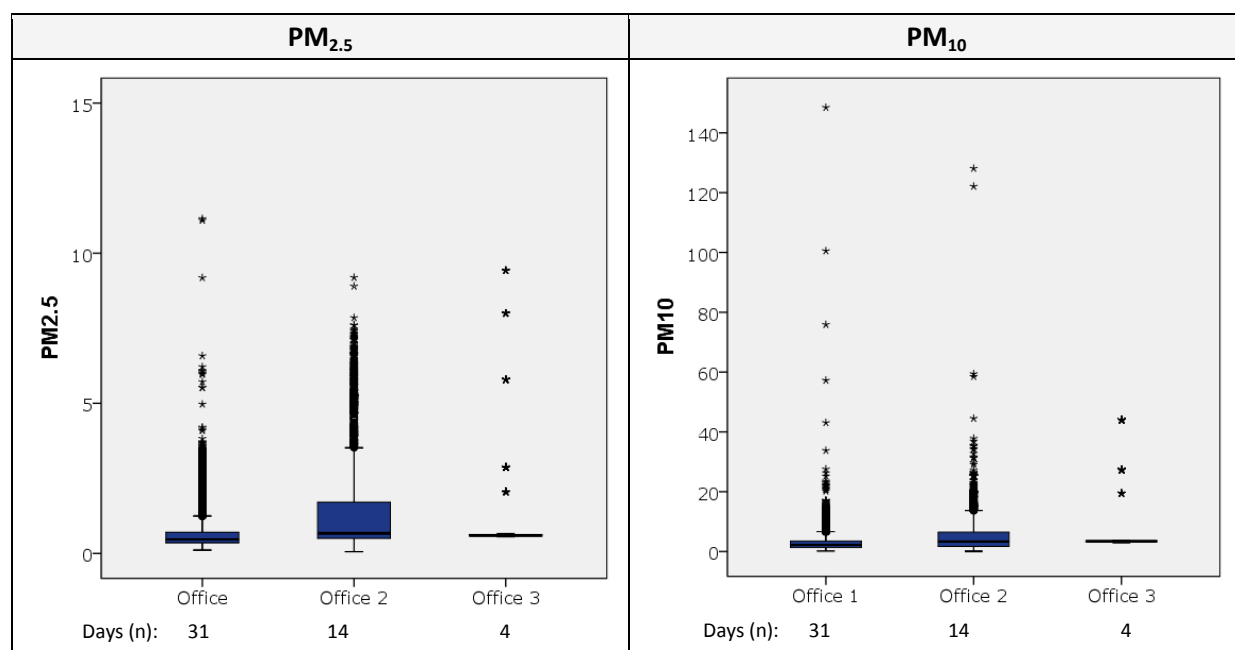


Figure 20: PM_{2.5} and PM₁₀ minute concentrations (both in µg/m³) recorded in different office rooms during personal exposure monitoring.

	n	PM _{2.5}		PM ₁₀	
	Days (Minutes)	Median (Ar. Mean)	IQR (SD)	Median (Ar. Mean)	IQR (SD)
Office	48 (21470)	0.55 (0.86)	0.43 (0.99)	2.75 (3.70)	2.31 (5.05)
Friend	1 (210)	16.74 (17.47)	9.25 (5.11)	43.38 (49.99)	36.48 (23.44)
Doctor	1 (39)	7.78 (8.27)	0.84 (1.63)	23.92 (31.31)	6.62 (38.65)
Nursery	2 (33)	4.01 (3.12)	2.48 (1.25)	25.20 (26.66)	15.25 (9.72)
Home A	39 (24,752)	4.31 (7.00)	3.21 (24.88)	11.20 (26.99)	10.50 (174.64)
Home B	2 (1,717)	4.14 (4.32)	2.40 (2.19)	9.60 (11.63)	3.70 (7.36)
Home D	4 (3,058)	4.50 (5.83)	3.00 (8.16)	8.60 (17.41)	8.10 (41.37)

Table 19: PM_{2.5} and PM₁₀ concentrations (both in µg/m³) of indoor microenvironments visited as part of the personal exposure monitoring campaign, based on minute concentrations.

Summary statistics of particle concentrations at indoor locations monitored during the personal exposure campaign are presented in Table 19. As little differences between office rooms were recognised, the office concentrations have been pooled for summary statistics. Results for the office

ME show very low median concentration of $0.55\mu\text{g}/\text{m}^3$ for $\text{PM}_{2.5}$ and $2.75\mu\text{g}/\text{m}^3$ for PM_{10} . Variability is as well very low with an IQR of $0.43\mu\text{g}/\text{m}^3$ for $\text{PM}_{2.5}$ and $2.31\mu\text{g}/\text{m}^3$ for PM_{10} . Median concentrations in home A in comparison are higher with $4.31\mu\text{g}/\text{m}^3$ for $\text{PM}_{2.5}$ and $11.20\mu\text{g}/\text{m}^3$ for PM_{10} . IQR for home A (indoors) are $3.21\mu\text{g}/\text{m}^3$ and $10.50\mu\text{g}/\text{m}^3$. For home D, results for $\text{PM}_{2.5}$ are similar to results for home A, with a median concentration of $4.50\mu\text{g}/\text{m}^3$ and an IQR of $3.00\mu\text{g}/\text{m}^3$. Results for PM_{10} concentrations are slightly below results for home A with a median of $8.60\mu\text{g}/\text{m}^3$ and an IQR of $8.10\mu\text{g}/\text{m}^3$ (For individual B not enough indoor monitoring concentration was recorded during personal monitoring for inclusion in the text, for individuals C no personal monitoring data was collected at home).

Apart from the homes and shared office location introduced above, three additional indoor locations have been visited once or twice during personal monitoring: a doctor's practice, and a nursery, as well as a friend's apartment. All three are naturally ventilated. Both the doctor's practice and the nursery are on the ground floor and were visited during opening hours. The friends' home is a first floor apartment and during the visit three other individuals were present (for basic statistics, see Table 19).

3.4.4. Bus Routes

Bus journeys were undertaken by all four individuals as part of their commute to work. The routes are named after the different individuals (routes A = individual A, routes B = individual B, etc.) Individual A took two buses to work, which alter their routes in some places on the way back. They are therefore presented here as four different bus routes (A1 – A4, numbered in the sequence they are taken during a day). Individuals B, C, and D took one bus route each (individual B and D in both directions, individual C in one direction). All buses run predominantly along major roads. The bus route taken by individual C was however only taken once, results are therefore not discussed.

In order to monitor concentrations in a bus a Dustmate monitor in a small rectangular bag was used for recordings during bus commutes. The monitor was stabilised in the bag by polystyrene, the inlet was sticking out at the top of the bag through a custom-cut hole. While standing in the bus, the bag was held in the hand (at least above knee height, usually higher), when seated in the bus, the bag was placed on the lap or on the neighbouring seat. The locations of the individuals in the buses varied. All buses were Diesel buses.

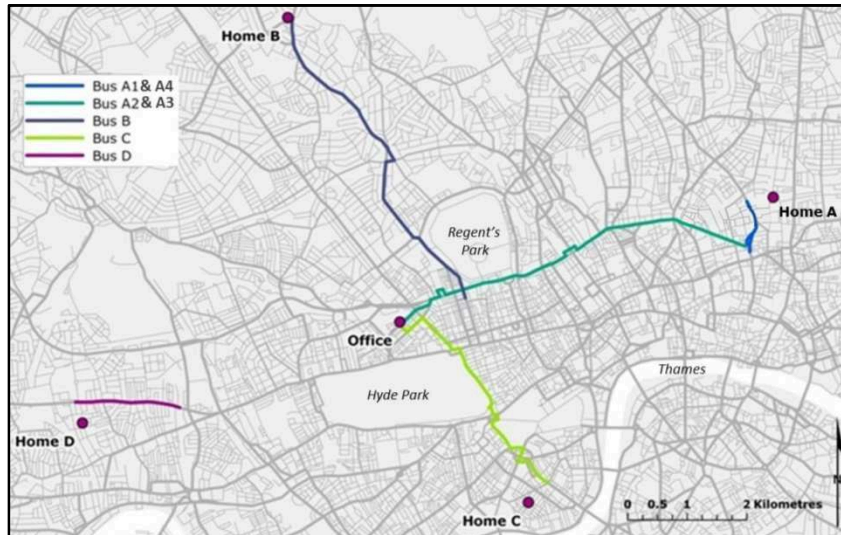


Figure 21: Central London, showing bus routes taken during personal exposure monitoring. Road Data Source: Meridian.

Summary statistics are presented in Table 20 for $PM_{2.5}$ and PM_{10} . The highest median concentrations were recorded on route A1 for both particle size fractions with median $PM_{2.5}$ concentrations of $16.61\mu g/m^3$ and median PM_{10} concentrations of $69.63\mu g/m^3$. Route A4 is the same bus route number as route 1, but taken on the return journey (with about $\frac{1}{4}$ along a different road in a one way system). Results for A4 show much lower concentrations compared to A1, as well as compared to the other routes. For $PM_{2.5}$ the median concentration for route A4 is $9.89\mu g/m^3$, and for PM_{10} the median concentration is $30.55\mu g/m^3$. The IQR are also lower in the evening compared to the morning, especially for PM_{10} (route A1 = $55.84\mu g/m^3$, route A4 = $32.46\mu g/m^3$). A potential explanation could be the increased traffic or the increased passenger number during the more pronounced morning rush-hour. According to Abt et al. (2000) the concentration increase from people's movement is more pronounced with larger particles, as it was observed here.

		Main Individual				Indiv. B	Indiv. C	Indiv. D
		Bus A1	Bus A2	Bus A3	Bus A4	Bus B	Bus C	Bus D
No. of repeats (minutes)		16 (65)	16 (668)	14 (572)	12 (41)	2 (123)	1 (17)	4 (83)
$PM_{2.5}$	Median (Mean)	16.61 (18.30)	12.21 (13.50)	8.27 (10.46)	9.89 (10.53)	5.98 (6.05)	13.92 (13.01)	3.87 (4.46)
	IQR (SD)	8.50 (11.34)	8.90 (8.18)	5.65 (7.99)	7.78 (6.21)	1.84 (1.42)	7.66 (5.14)	2.15 (2.31)
PM_{10}	Median (Mean)	69.63 (83.38)	43.45 (53.54)	31.96 (39.31)	30.55 (43.27)	23.20 (24.35)	56.40 (51.85)	19.80 (21.75)
	IQR (SD)	55.84 (54.28)	35.31 (52.71)	30.85 (6.37)	32.46 (36.19)	7.40 (6.63)	29.65 (21.04)	11.10 (8.24)

Table 20: $PM_{2.5}$ and PM_{10} concentrations (both in $\mu g/m^3$) of bus routes visited as part of the personal exposure monitoring campaign, based on minute concentrations

Bus A2 and A3 are as well the same bus route (with one-way alterations) taken in different directions, with A2 is taken during morning rush hour, and A3 taken in the evening. As for routes A1 and A4, PM concentrations are higher in the morning, but the difference is not as pronounced as along route A1/A4. For PM_{2.5} median levels in the morning is 12.21µg/m³ compared to 8.27µg/m³ in the afternoon; median PM₁₀ concentrations in the morning is 43.45µg/m³ and only 31.96µg/m³ in the afternoon.

Route A2/A3 runs along some of the busiest roads in London (Euston Road, Marylebone Rd. etc.) with DAT of on average around 39,000 (length weighted average along LAEI road segments). Along routes A1/A4 in comparison DAT is just above 15,000. Several studies have shown that in-vehicle concentrations on high traffic roads are higher than on low traffic roads (Vijayan & Kumar 2010; Zuurbier et al. 2010). Despite the higher DAT, routes A2/A3 show however lower particle concentrations. Other factors therefore may be of importance, such as window openings or passenger numbers (see chapter 2). Concentrations for bus B are the second lowest recorded along the different bus routes. The median PM_{2.5} concentration is 5.98µg/m³ and median PM₁₀ is 23.2µg/m³. Additionally the IQR are low, and show only 1.84µg/m³ for PM_{2.5} and 7.4µg/m³ for PM₁₀. Individual B chose however to sit upstairs on all journeys, which may potentially have had a reduction effect on the PM levels due to vertical displacement. Bus D has the lowest median values with median PM_{2.5} concentration of only 3.87µg/m³ and of 19.8µg/m³ for PM₁₀. The DAT along the route is also one of the lowest compared to other bus routes with DAT of just above 15,000 (weighted by segment lengths).

Several other studies have collected data during bus journeys, generally finding higher concentrations than described above for this thesis. Mean PM_{2.5} concentrations of in-bus studies presented in chapter 2 lie between 34.5µg/m³ and 128.16µg/m³ for in-bus exposure (Adams et al. 2001; Kaur et al. 2005b; McNabola et al. 2009a; Zuurbier et al. 2010). In comparison mean PM_{2.5} concentrations presented in Table 20 lie between 4.46µg/m³ and 18.30µg/m³. Only two studies provide results for PM₁₀ inside buses. One study by Praml & Schierl (2000) also collected PM₁₀ concentrations inside buses, finding mean concentrations of 110.08µg/m³ and 164.95µg/m³ for two different buses in Munich, Germany. Again, results by this thesis are lower with a mean concentration of between 21.75µg/m³ and 83.38µg/m³ (see Table 20). Results from a study, by Zuurbier et al. (2010) show median PM₁₀ concentration of 46.7µg/m³ for a Diesel bus route. These findings lie below the median concentration for route A1, but higher than median concentrations for all other routes in this thesis. Many explanations for the difference to other studies are possible as in-bus concentrations change with location and time, and different study set ups may have captured different ranges of concentrations. Conclusions are therefore difficult.

3.4.5. Cycling Routes

All cycling during personal monitoring was undertaken by individual A. Route locations are presented in Figure 22 (left side). In order to record air pollution during cycling routes, a special set up was built for an OSIRIS monitor (see Figure 22, right side). The monitor was placed in a front basket of the bicycle on top of a cushioning foam layer; it was held in place by bicycle extension straps. The inlet was facing towards the front. All cycling was undertaken in central London, where cyclists share the lanes with cars and buses.

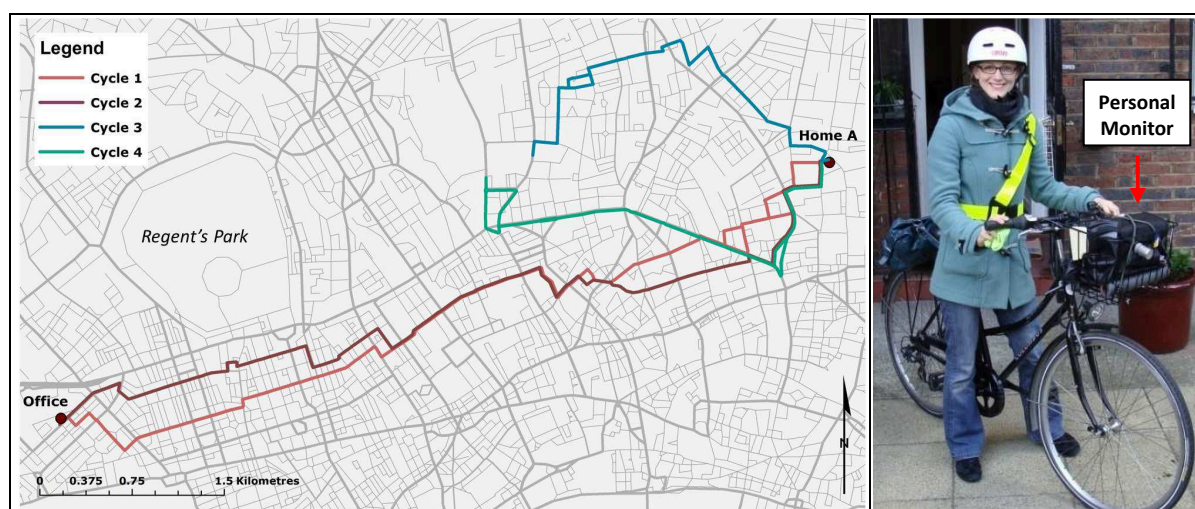


Figure 22: Left side: central London, showing cycling routes taken during the personal exposure monitoring campaign. Road data source: Meridian. Right side: Cycle monitor set up.

Summary statistics are presented in Table 21 including average traffic along the routes, calculated as length weighted averages from LAEI data (per LAEI road segment). Smaller roads are not included in the LAEI dataset (e.g. most of route 3). Approximate percentage of LAEI data coverage along the routes is added therefore to the table.

Route 1 and 2 follow along parallel routes. Route 1 is the route from home to work, route 2 the route from work back home. They experience similar medium high average traffic levels (compared to the other cycling routes). Both routes avoid major roads where possible; they lead however through some of the busiest parts of the centre of London. For most of the routes DAT is with about 9,000 slightly higher on route 1 than on route 2 with 8,500. Monitoring on route 1 was additionally undertaken in the morning peak traffic hours, whereas route 2 was monitored in the evenings. Both reasons (DAT difference and time of day) might explain the higher median and IQR along route 1 compared to route 2. For $PM_{2.5}$ the median concentration on route 1 is $10.82\mu g/m^3$ and the IQR is $10.2\mu g/m^3$ compared to a median of $6.78\mu g/m^3$ and an IQR of $7.33\mu g/m^3$ on route 2. For PM_{10} the

median on route 1 is 39.63 $\mu\text{g}/\text{m}^3$ and the IQR is 33.76 $\mu\text{g}/\text{m}^3$; for route 2 the median is 27.98 $\mu\text{g}/\text{m}^3$ and the IQR is 25.61 $\mu\text{g}/\text{m}^3$.

		Cycle 1	Cycle 2	Cycle 3	Cycle 4
No. of days (minutes)		23 (833)	24 (877)	1 (86)	1 (123)
PM _{2.5}	Median (Ar. Mean)	11.05 (16.53)	7.41 (10.69)	5.32 (5.26)	16.22 (17.95)
	IQR (SD)	10.96 (22.29)	7.14 (11.65)	1.83 (1.37)	9.63 (9.67)
PM ₁₀	Median (Ar. Mean)	39.95 (57.20)	29.95 (39.27)	20.34 (20.80)	65.15 (69.78)
	IQR (SD)	36.71 (66.31)	27.78 (39.15)	8.74 (6.76)	29.30 (27.49)
DAT (Approx. traffic data coverage)		9,197 (80% cov.)	8,457 (80% cov.)	5,481 (20% cov.)	18,933 (90% cov.)

Table 21: PM_{2.5} and PM₁₀ concentrations (in $\mu\text{g}/\text{m}^3$ and based on minute data) of cycling routes visited as part of the personal exposure monitoring campaign, as well as daily average traffic (DAT). Traffic data source: LAEI.

Concentrations recorded by other studies for cycling exposure show generally higher results than described above for this thesis. As described in the chapter 2 mean concentrations for PM_{2.5} in the literature range between 23.5 $\mu\text{g}/\text{m}^3$ and 88.14 $\mu\text{g}/\text{m}^3$ during cycling (Adams et al. 2001; Kaur et al. 2005b; McNabola et al. 2009a; Zuurbier et al. 2010). Mean cycling concentrations collected during personal monitoring for this thesis lie on the other hand only between 5.26 $\mu\text{g}/\text{m}^3$ and 17.95 $\mu\text{g}/\text{m}^3$ (see Table 21). The study by Zuurbier et al. also recorded PM₁₀ concentrations during cycling and recorded median concentrations of 37 $\mu\text{g}/\text{m}^3$ and 39.3 $\mu\text{g}/\text{m}^3$ on two different routes. These two median concentrations lie in between median results for cycling routes recorded during personal monitoring for this thesis (between 20.34 $\mu\text{g}/\text{m}^3$ and 65.15 $\mu\text{g}/\text{m}^3$). Again, as for studies on in-bus concentrations, it is difficult to draw any conclusions from the comparison, as other studies differ in time and location, as well as in set-up and purpose.

3.4.6. Walking Routes

All four individuals spent time walking along at least two different routes during personal monitoring. The locations of the routes are shown in Figure 23. Most walking routes have however been only visited during short time periods or for little or no repeats. As before, only results for locations that have been visited at least three times are described here.

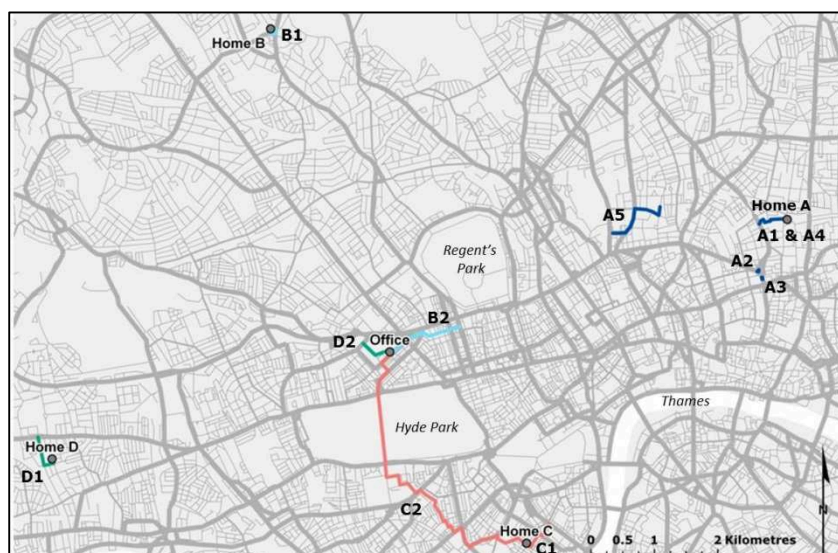


Figure 23: Central London, showing walking routes during the personal exposure monitoring campaign. Road data source: Meridian.

During walking routes the personal monitor was, as during bus routes, carried in the custom-made bag. It was possible to wear the bag around the neck or around the neck and one arm. In both cases the bag was carried at the front of the body and above hip height.

Walking exposure is directly determined by ambient particle concentrations. There is however a certain element of displacement from the traffic sources along the routes due to the position on the sidewalk. Depending on factors like road width and wind speed, the influence of the local traffic component can be altered (Kaur et al. 2005b). Of the recorded walking routes, some follow only roads with little traffic: A1, A4, D1 and D2; some follow along a routes that vary in the amount of traffic: A5, B1, B2, C1, and C2; and some routes only follow along high traffic roads: A2 and A3 (both at Old Street Roundabout: traffic varying between about 11,000 and 46,000 DAT).

Summary statistics of the different walking routes are presented in Table 22. The highest median values of the walking routes have been recorded by individual A along the four short routes of A1, A2, A3, and A4. A2 and A3 have been recorded during the morning commute, A3 and A4 have been recorded during evening commute (A1 and A4 are otherwise the same). As observed for some of the bus routes above, the morning rush-hour appears to cause much higher concentrations. The $PM_{2.5}$ median concentration for route A1/A4 is $10.08\mu g/m^3$ in the morning and $6.92\mu g/m^3$ in the evening. The median PM_{10} value on route A1 is $43.88\mu g/m^3$ and only $28.05\mu g/m^3$ in the evening on route A4. Similarly, for route A2/A3 the morning median concentration (A2) is higher than in the evening (A3) for $PM_{2.5}$ (A2: $12.15\mu g/m^3$, A3: $10.01\mu g/m^3$) and for PM_{10} (A2: $65.58\mu g/m^3$, A3: $36.00\mu g/m^3$).

		Main Individual					Individual B		Individual C		Individual D	
		A1	A2	A3	A4	A5	B1	B2	C1	C2	D1	D2
No. of days (min.)		22 (120)	14 (33)	12 (35)	11 (72)	1 (13)	2 (38)	2 (19)	1 (12)	2 (147)	4 (47)	3 (34)
PM _{2.5}	Median (Mean)	10.08 (11.38)	12.15 (17.56)	10.01 (10.84)	6.92 (7.90)	5.90 (6.81)	5.06 (5.44)	4.12 (4.70)	4.65 (6.55)	4.02 (4.26)	3.45 (4.16)	2.77 (2.95)
	IQR (SD)	6.35 (6.39)	7.91 (19.25)	6.96 (4.66)	6.03 (5.10)	1.39 (3.64)	1.75 (1.43)	1.63 (1.93)	6.85 (3.71)	2.99 (2.97)	4.09 (2.61)	0.96 (1.39)
PM ₁₀	Median (Mean)	43.88 (55.70)	65.58 (90.56)	36.00 (63.06)	28.05 (34.42)	29.05 (140.52)	25.10 (27.31)	16.10 (18.89)	18.65 (26.18)	18.40 (19.89)	18.50 (22.94)	14.75 (18.85)
	IQR (SD)	25.76 (51.81)	34.50 (140.89)	33.19 (82.16)	27.34 (26.63)	36.70 (331.05)	8.40 (7.68)	10.00 (7.80)	25.33 (14.76)	12.90 (13.78)	11.20 (15.15)	7.45 (16.03)

Table 22: PM_{2.5} and PM₁₀ concentrations (both in $\mu\text{g}/\text{m}^3$) along walking routes, collected during the personal exposure monitoring campaign, based on minute data.

Both routes of individual D are background routes and for all three size fractions they show some of the lowest median concentrations with a median of $3.45\mu\text{g}/\text{m}^3$ (D1) and $2.77\mu\text{g}/\text{m}^3$ (D2) for PM_{2.5}, and medians of $18.50\mu\text{g}/\text{m}^3$ (D1) and $14.75\mu\text{g}/\text{m}^3$ (D2) for PM₁₀.

There exist several studies in the literature that have collected concentrations during walking routes, as described in chapter 2. For PM_{2.5} mean concentrations of different studies on walking exposure lie between $6.59\mu\text{g}/\text{m}^3$ and $63.45\mu\text{g}/\text{m}^3$ (Briggs et al. 2008; Gulliver & Briggs 2004; Gulliver & Briggs 2007; Kaur et al. 2005b; Kaur et al. 2005a; McNabola et al. 2009a). Mean concentrations of results presented above as part of this thesis fall into the lower part of this range with between $2.95\mu\text{g}/\text{m}^3$ and $17.56\mu\text{g}/\text{m}^3$ (see Table 22). For PM₁₀ mean concentrations from the literature lie between $22.1\mu\text{g}/\text{m}^3$ and $38.18\mu\text{g}/\text{m}^3$, which is relatively low compared to the range of mean concentrations of between $18.85\mu\text{g}/\text{m}^3$ and $140.52\mu\text{g}/\text{m}^3$ in this study (see Table 22). As for the results of other traffic MEs, differences between studies may be due to differences in time and space, as well as study set ups.

3.4.7. Subway Route

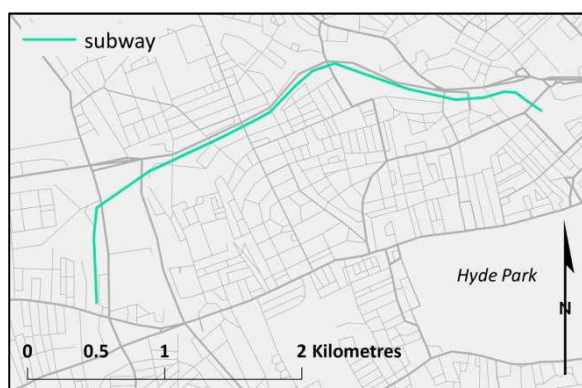


Figure 24: Central London, showing the location of a subway route taken as part of the personal exposure monitoring campaign. Road data source: Meridian

Additionally to the transport MEs described above, some personal monitoring was undertaken in the subway system along one route (location presented in Figure 24). The subway route was only taken by individual D during four consecutive days (eight rides). The section of the subway route belongs to the Hammersmith and City line and is (apart from the stations) entirely above ground.

Basic statistics are presented in Table 23 showing a median concentration of $10.59\mu\text{g}/\text{m}^3$ and an IQR of $12.66\mu\text{g}/\text{m}^3$ for $\text{PM}_{2.5}$ and a median concentration of $45.20\mu\text{g}/\text{m}^3$ and an IQR of $50.15\mu\text{g}/\text{m}^3$ for PM_{10} .

	n	PM _{2.5}			PM ₁₀		
	Days (Minutes)	Median (Ar. Mean)	IQR (SD)	95 th Percentile	Median (Mean)	IQR (SD)	95 th Percentile
Subway	4 (77)	10.59 (13.88)	12.66 (13.46)	44.70	45.2 (63.06)	50.15 (66.53)	194.38

Table 23: $\text{PM}_{2.5}$ and PM_{10} concentrations for times in the subway system, collected during the personal exposure monitoring campaign, based on minute concentrations.

Local trains above ground have shown much lower particle concentrations compared to underground trains, even within the same subway system. A study by Hurley et al. (2004) for example shows for $\text{PM}_{2.5}$ concentrations in the London underground system that concentrations collected when under ground along the same subway line are between 4.74 and 10.9 times higher than during times at the surface.

Few studies have presented concentrations specifically for over-ground subways or local trains. Results from the study by Hurley et al. (2004) can be calculated into mean concentrations for above-ground times (from average concentrations per line and ratio below/above ground, if equal time above and below ground are assumed). Mean concentrations for times above ground are as such $12.12\mu\text{g}/\text{m}^3$ for the Central line, $26.04\mu\text{g}/\text{m}^3$ for the Northern line and $23.74\mu\text{g}/\text{m}^3$ for the Victoria line. The mean concentration of $13.88\mu\text{g}/\text{m}^3$ from this thesis falls within that range. No results for PM_{10} could be found in the literature specifically for concentrations in subway systems above

ground. Results by (Praml & Schierl 2000) for trams however show mean concentrations of $183.41\mu\text{g}/\text{m}^3$, which is almost three times the mean concentration observed for the subway route of $63.06\mu\text{g}/\text{m}^3$.

3.5. Transport Monitoring Campaigns

3.5.1. Introduction and Set Up of Transport Monitoring Campaigns

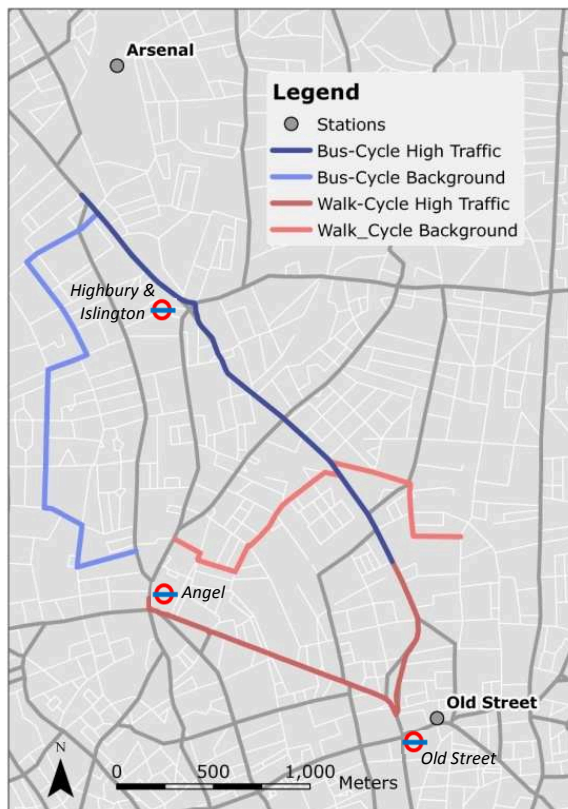


Figure 25: Central London (Islington): routes visited as part of the transport monitoring campaign. Road data source: Meridian

Simultaneous monitoring of transport modes was undertaken in order to collect data that can be used to calculate adjustments for personal exposure in traffic MEs. Data was collected for this purpose during two bespoke monitoring campaigns: 1) A walking-cycling campaign and 2) a bus-cycling campaign. The way the monitor was carried and set up in the different modes has been described above for personal monitoring. GPS data was collected during all runs in order to locate recorded concentrations for both modes along the routes; details and times of each run were noted in a diary. A background monitor of the same type as the campaign monitors (Osiris) was installed at Kensington and Chelsea - North Kensington (location: see Figure 10), a central London background station during 5 of the 7 monitoring days (installation had to be delayed by two days for access reasons).

During the two weeks of monitoring, two particle air pollution episodes had been recorded for London by the LAQN (Early March 2010: 2nd – 9th, Mid-March 2010: 15th – 17th), which overlapped with the monitoring times (7th to 15th of March 2011). Concentrations collected during this campaign were most likely influenced by increased ambient concentrations during the episodes.

Set up and Route Description for Walking-Cycling Comparison:

PM concentrations for walking and cycling were simultaneously recorded 10 times on one low traffic route and 8 times on one high traffic route (see Figure 25). Two routes with different traffic counts were chosen to evaluate differences between the walking to cycling relationship for different types of routes. The low traffic route has a length of 2.2km and generally follows quiet streets apart from the crossing of one major road. Traffic counts are not available for most of the route due to low traffic numbers. The high traffic route is 2.1km long and runs along major roads the whole way. Traffic counts are available on all road segments and the average length weighted traffic count is 21,026 DAT. During monitoring, walker and cyclist started at the same time at the same point and, on completion of the journey, the cyclist waited at the end of the route (being about twice as fast). The route was always recorded in one direction. The cyclist was always cycling in the car or bus lane (no cycling path existed along any of the routes) and the walker took care to use the sidewalk on the same side of the street (left). Position on the sidewalk has been shown to make an important difference in exposure, as concentrations drop within few meters away from traffic sources (Kaur et al. 2005a). The walker was therefore advised to walk in the middle of the sidewalk where possible.

Set up and Route Description for Bus-Cycling Comparison:

A bus-cycling comparison was undertaken along one low traffic route and one high traffic route (see Figure 25). Two routes with different traffic counts were chosen to evaluate differences between the cycling to in-bus relationship for different types of routes. Each route was simultaneously monitored for 15 times on the low traffic route and for 17 times on the high traffic route. The low traffic route was 2.8km in length. Most of the route has very low traffic counts and is therefore not included in the traffic count data by the LAEI. For about 1/3 of the route the daily average traffic is available with an average of 5,525. The high traffic route is 2.24km long, and an average (length-weighted) number of 21,644 vehicles pass it per day.

During monitoring both volunteers started from the same bus stop, the cyclist waited in front of the stop until the bus started moving. The cyclist then stayed just in front of the bus (up to about 100m) during the whole journey, slowing down to wait for the bus during bus halts. In only few occasions the bicycle actually had to stop fully to wait for the bus. Keeping close proximity between the modes was preferred, as this reduces variation due to different local transport influences. This set up (bicycle in the front) was chosen to avoid direct influences of the bus' exhaust on the cyclist's exposure. Only at two occasions during all runs did the bus overtake the cyclist and stayed in front

for a couple of minutes. The volunteer in the bus was located on the lower deck during all repeats and was sitting down on most journeys. All buses were double-decker buses, powered by diesel.

3.5.2. Description of Data Collected during Transport Monitoring Campaigns

Results of Simultaneously Monitored Walking and Cycling MEs:

Results for the different routes and modes (see Table 24) show that for both particle size fractions, as well as for both route types, the median concentration is higher for the cyclist than for the walker. Between the route types (low and high traffic) however only small differences can be observed for $PM_{2.5}$; for PM_{10} median concentrations along the low traffic route are higher compared to the high traffic route:

Along the low traffic route median concentrations for $PM_{2.5}$ are $9.91\mu g/m^3$ for walking and $18.79\mu g/m^3$ for cycling. On the high traffic route, median $PM_{2.5}$ values are $9.48\mu g/m^3$ for walking and $19.76\mu g/m^3$ for cycling.

For PM_{10} median concentrations on the low traffic route are $41.40\mu g/m^3$ for walking and $48.50\mu g/m^3$ for cycling. Median PM_{10} concentrations on the high traffic route are $36.70\mu g/m^3$ for walking and $49.70\mu g/m^3$ for cycling.

As the cyclist is closer to the highly variable traffic source influences, the variability was initially expected to be also higher for cycling than for walking. For $Pm_{2.5}$ the IQR on the low traffic route the IQR is $21.35\mu g/m^3$ for cycling, and $18.04\mu g/m^3$ for walking. Along the high traffic route the IQR is $18.1\mu g/m^3$ for cycling compared to only $6.29\mu g/m^3$ for walking.

For PM_{10} the IQR on the low traffic route is as well higher for cycling with $41.57\mu g/m^3$ for cycling and $31.42\mu g/m^3$ for walking. Along the high traffic route walking shows higher variability with an IQR of $27.4\mu g/m^3$ compared to $23.68\mu g/m^3$ for cycling. No direct explanation for the higher variability for some size fractions on the low traffic route can be given. Some potential explanations could lie in local winds or other non-traffic related influences.

	PM _{2.5}						PM ₁₀					
	Low traffic			High traffic			Low traffic			High traffic		
	Walk	Cycle	BG	Walk	Cycle	BG	Walk	Cycle	BG	Walk	Cycle	BG
Minutes (journeys)	256 (10)	104 (10)	196	235 (8)	136 (8)	181	256 (10)	104 (10)	196	235 (8)	136 (8)	181
Median (Mean)	9.91 (19.24)	18.79 (25.40)	6.21 (18.40)	9.48 (14.18)	19.76 (24.49)	8.09 (11.90)	41.40 (47.84)	48.50 (61.22)	16.10 (28.31)	36.70 (45.35)	49.70 (57.96)	17.30 (22.30)
IQR (SD)	18.04 (19.2)	21.35 (22.16)	25.04 (2019)	6.29 (13.45)	18.1 (15.99)	3.99 (12.78)	31.42 (36.03)	41.57 (49.02)	30.73 (21.87)	27.4 (30.85)	23.68 (41.21)	13.00 (14.06)
99th P.	73.18	90.64	72.44	55.81	68.63	39.02	161.00	295.70	86.28	188.68	313.11	50.53

Table 24: PM_{2.5} and PM₁₀ concentrations (both in µg/m³ and based on minute concentrations) for simultaneous walking and cycling monitoring along a high traffic and a low traffic route, as well as for stationary monitoring concentrations at a background location. BG = stationary background monitor.

Concentrations for the stationary background monitor (BG) are as well given for times of monitoring along the different routes. Selection of minutes from the BG monitor is based on times of walking (which took longer than cycling). Results for the BG monitor show lower median concentrations compared to either transport mode, for both routes and for PM_{2.5}, as well as PM₁₀. Median concentrations for PM_{2.5} are 6.21µg/m³ during low traffic route monitoring and 8.09µg/m³ during high traffic route monitoring. For PM₁₀, median concentrations at the BG station are 16.10µg/m³ during low traffic route monitoring, and 17.30µg/m³ during high traffic route monitoring.

Compared to differences between modes, the differences observed between route types (low traffic and high traffic) are small. For walking the median concentrations are higher on the low traffic route where the individual should be exposed to less traffic sources. A higher variability would be expected on the high traffic route, due to the proximity of many (moving) sources. However, the IQR is lower on the high traffic route for all modes, route types, and for both particle size fractions. For walking one potential explanation for the lower concentrations along the high traffic route could be the distance from the kerb. The roads along the high traffic route, including the pavement, are in many places much wider than along the low traffic route. This might have resulted in a greater distance from the sources. Additionally, the chosen high traffic route provides much more open space around the road area aiding fast dispersion of particles.

In comparison to the walking and cycling routes monitored during personal monitoring (presented above), some of the median concentrations, as well as some of the IQR are higher in this walking-cycling transport campaign. The reason for this may however lie in the pollution episode during this traffic monitoring campaign.

Results of Simultaneously Monitored Bus and Cycling MEs:

Results for the bus-cycling campaign are presented in Table 25. $PM_{2.5}$ median values are higher for cycling than for in-bus concentrations. $PM_{2.5}$ concentrations show on the low traffic route a median of $14.70\mu g/m^3$ for cycling and $14.03\mu g/m^3$ for inside the bus. Along the high traffic route the median cycling concentration is $15.68\mu g/m^3$ and $14.03\mu g/m^3$ inside the bus. The IQR for $PM_{2.5}$ are as well higher for cycling than for bus. For $PM_{2.5}$ the in-bus IQR are $12.85\mu g/m^3$ (low traffic route) and $13.66\mu g/m^3$ (high traffic route) compared to $21\mu g/m^3$ (low traffic route) and $18.82\mu g/m^3$ (high traffic route) for cycling.

For PM_{10} , higher median concentrations are recorded inside the bus compared to cycling. On the low traffic route the median in the bus is $55.60\mu g/m^3$, on the high traffic route it is $66.30\mu g/m^3$. The cyclist only recorded median concentrations of $43\mu g/m^3$ on the low traffic route and $46.2\mu g/m^3$ on the high traffic route. The IQR is on the low traffic route however higher for cycling ($41.4\mu g/m^3$) than for the bus ($35.1\mu g/m^3$). On the high traffic route the IQR is with $38.7\mu g/m^3$ lower for cycling, compared to $52.7\mu g/m^3$ in the bus. Generally it can be assumed that, as described for home indoor environments, there is a certain amount of re-suspension of dust particles inside the bus cabin, caused by the density of people in a small room and their movements. It has also been found that resuspension is higher for larger particles (Abt, Catalano, et al. 2000; Long et al. 2001). This would provide an explanation for high PM_{10} concentrations and high variability inside the bus compared to the cyclist outside.

The monitor located at a stationary background location (BG) shows lower median concentrations compared to either transport mode on both routes and for $PM_{2.5}$, as well as for PM_{10} . Median $PM_{2.5}$ concentrations at the BG monitor are $6.98\mu g/m^3$ during monitoring of both routes. Median PM_{10} concentrations are $17.18\mu g/m^3$ and $16.68\mu g/m^3$ for times of monitoring along the low traffic and high traffic route respectively.

	PM _{2.5}						PM ₁₀					
	Low traffic			High traffic			Low traffic			High traffic		
	Bus	Cycle	BG	Bus	Cycle	BG	Bus	Cycle	BG	Bus	Cycle	BG
Minutes (journeys)	247 (15)	279 (15)	249	223 (17)	254 (17)	221	247 (15)	279 (15)	249	223 (17)	254 (17)	221
Median (Ar. Mean)	10.63 (17.47)	14.70 (23.32)	6.98 (13.77)	14.03 (19.19)	15.68 (23.60)	6.98 (11.60)	55.60 (63.47)	43.00 (60.82)	17.18 (23.67)	66.30 (81.90)	46.20 (62.99)	16.68 (21.40)
IQR (SD)	12.85 (13.92)	21.00 (21.94)	5.79 (15.15)	13.66 (13.45)	18.82 (19.16)	5.79 (12.73)	35.10 (27.26)	41.4 (94.61)	13.70 (16.28)	52.70 (42.46)	38.70 (73.03)	7.90 (14.07)
99th P.	59.89	76.66	50.30	58.78	89.00	50.30	143.56	393.62	60.35	245.91	366.86	60.35

Table 25: PM_{2.5} and PM₁₀ concentrations (both in µg/m³ and based on minute concentrations) for simultaneous bus and cycling monitoring along a high traffic and a low traffic route, as well as for stationary monitoring concentrations at a background location. BG = Stationary background monitor.

Another observation from this thesis is that both modes show slightly higher median concentrations along the high traffic route compared to the low traffic route. For the cyclist this difference most likely reflects higher source activities along the high traffic route. For the bus, influences from inside the bus may have effected on these differences as well.

3.6. Co-Location of Osiris Monitors with Network Station Monitors

Co-location between Osiris monitors and TEOM/FDMS (tapered element oscillating microbalance/ Filter Dynamics Measurement System) monitors took place at two permanent monitoring stations of the London Air Quality Network (LAQN) between March and May 2011. Co-locations took place at Westminster - Marylebone station (ML), a traffic location, for 42 (PM_{2.5}) and 41 (PM₁₀) days, and at Kensington and Chelsea - North Kensington station (NK), a background location, for 16 days (locations are presented in Figure 10). Monitors for PM_{2.5} at the two stations are FDMS devices; PM₁₀ is monitored with TEOM monitors. Data for the FDMS/TEOM monitors at the stations were provided online by the LAQN (<http://www.londonair.org.uk/london>, accessed: 28th July 2011). An additional dataset was kindly provided by Shukri Mohd from King's College, who had co-located an Osiris monitor with TEOM monitors (both for PM_{2.5} and PM₁₀) at an urban background location in Greenwich for 37 days during March/April 2010.

Basic statistics of concentrations from Osiris and FDMS/TEOM monitors for all three locations are presented in Table 26. Concentrations are presented at 15 minute intervals, as this was the highest available temporal resolution for concentrations for data from the network monitoring stations. Results for PM_{2.5} show that at all locations the median, IQR and range were lower for the Osiris monitors with median concentrations between 6.30µg/m³ and 9.58µg/m³ for the Osiris and median concentrations of between 17.00µg/m³ and 29.00µg/m³ for the FDMS/TEOM monitors. IQR results

for the Osiris monitors range between $3.80\mu\text{g}/\text{m}^3$ and $7.81\mu\text{g}/\text{m}^3$. Results for IQR recorded by FDMS/TEOM monitors are much higher with between $9.00\mu\text{g}/\text{m}^3$ and $32.00\mu\text{g}/\text{m}^3$. For PM_{10} median, IQR and range are as well lower for concentrations recorded by the Osiris monitors, compared to the TEOM devices at each location. Results for median concentrations range between $15.40\mu\text{g}/\text{m}^3$ and $33.50\mu\text{g}/\text{m}^3$ for concentrations recorded by the Osiris and between $23.20\mu\text{g}/\text{m}^3$ and $45.00\mu\text{g}/\text{m}^3$ recorded by TEOM monitors. The IQR ranges between 9.80 and 19.10 for recordings by the Osiris monitors, and between $12.60\mu\text{g}/\text{m}^3$ and $29.00\mu\text{g}/\text{m}^3$ for recordings by the TEOM monitors.

	$\text{PM}_{2.5}$						PM_{10}					
	ML		NK		GR		ML		NK		GR	
	Osiris	FDMS	Osiris	FDMS	Osiris	TEOM	Osiris	TEOM	Osiris	TEOM	Osiris	TEOM
15min intervals	3788	3788	1832	1832	3349	3349	3788	3788	1832	1832	3349	3349
Median (Mean)	9.58 (11.62)	24.00 (28.14)	9.12 (13.02)	29.00 (32.69)	6.30 (7.01)	17.00 (18.25)	33.50 (35.71)	45.00 (49.01)	20.30 (24.32)	36.00 (40.06)	15.40 (17.54)	23.20 (25.41)
IQR (SD)	6.73 (7.69)	21.00 (18.66)	7.81 (11.88)	32.00 (18.94)	3.80 (3.47)	9.00 (7.57)	19.10 (16.07)	29.00 (22.28)	15.98 (14.13)	28.00 (19.37)	9.80 (8.71)	12.60 (10.56)
Range	57.65	107.00	81.28	83.00	21.80	42.00	135.60	270.00	93.40	128.00	71.50	54.50

Table 26: $\text{PM}_{2.5}$ and PM_{10} concentrations at 15-minute intervals for TEOM/FDMS monitors and co-located Osiris monitors at Westminster – Marylebone (ML) station, at Kensington and Chelsea – North Kensington (NK) station, and at Greenwich station (GR).

The presented results clearly show underprediction of the Osiris monitors in comparison to EU reference equivalent monitoring methods, FDMS/TEOM, both for $\text{PM}_{2.5}$ and PM_{10} at all three locations. For the analyses in this thesis concentrations monitored by Osiris devices, as well as concentrations recorded by EU reference equivalent monitors are used. Calibrations of Osiris monitors to EU reference equivalence were therefore considered necessary prior to analyses. The calibration method is presented in Appendix F.

3.7. Preparing Monitored Particle Concentrations for Analyses

3.7.1. Data Preparation

Outlier Removal

Monitored datasets as described in this chapter showed some very high concentration values at minute level, for example during cooking events or in traffic situations. High concentrations even during short time periods contribute to adverse health effects caused by particle exposure. High

values on the other hand can provide a mathematical problem, as they can influence for example the mean concentration of an hourly average or the fit of a model when compared to monitoring data. In order to avoid these effects, high (or low) concentrations that are very different to the rest of the data distribution should therefore be removed as outliers.

As preparation for the analyses in this thesis it was decided to remove unusual values. Outlier removal was undertaken at a minute level to a) allow for a more precise removal of single values, and b) avoid unnecessary removal of data points. There are several common methods for outlier removal, such as Chauvenet's criterion or Grubbs' test. They however assume normal distribution of a dataset, which is not met by the monitored data of this thesis. Some other methods focus on removing single outliers. Datasets collected for this theses however frequently showed several very high values. A generic removal of extreme concentrations is sometimes practiced, such as removal of concentrations outside a range of twice the IQR around the mean. This approach is however not feasible for the datasets of this thesis as it would remove many high concentrations during and after cooking, as well as peak concentrations in traffic. The best remaining method was the removal of outliers from visual observation using box plots, as suggested by Harris & Jarvis (2011). Visual removal of outliers relies on visually observing gaps between the majority of data points and data points that are much above or below most other values. Removal of outliers for this thesis was based on the main principle of visual observation. In order to assure consistency within this thesis the following protocol was however followed:

- Concentrations within twice the IQR of a dataset are never removed as outliers.
- For concentrations above two times the IQR (no low extreme values existed in any of the datasets) the following rules were observed:
 - For datasets with a range below $1000\mu\text{g}/\text{m}^3$: If the distance between two data values is more than 10% of the dataset's range, all minute concentrations above this 'gap' in the dataset are removed.
 - For datasets with a range above $1000\mu\text{g}/\text{m}^3$: If the distance between two data values is more than 5% of the dataset's range, all minute concentrations above this 'gap' in the dataset are removed.

Removed data values are indicated in the data distributions presented in Appendix G.

In a few of the following analyses, extreme data values at an hourly level substantially altered results. In these occasions it was decided to remove the extreme values in the same way described above for minute concentrations. Results for these analyses are however always presented both with and without extreme values.

Addressing Skewed Data Distributions

Datasets of particle concentrations are often positively skewed, as they have a minimum level of zero and no upper limit (Kruize et al. 2003). A positively skewed dataset has a tail of high values away from the mean (Harris & Jarvis 2011). Several methods applied in the following analyses, such as Pearson's r and R^2 , are influenced by the skewness of a dataset. Particle concentrations with high temporal frequency, such as minutes, show often particularly high skewness, as the variability of minute concentrations can be high. All datasets described above have been to some extent aggregated for the following analyses. After aggregating the datasets and removal of minute outliers many of the datasets were approximately normally distributed with a skewness factor of between -1 and 1 (Harris & Jarvis 2011).

For the remaining positively skewed datasets, a potential method to reduce skewness would be by log transformation. It was however decided against log transforming skewed datasets, as log transformation would reduce the importance of extreme values in an analysis. High concentrations are however of particular importance in human exposure to particles. Reducing their importance in the analyses is therefore not considered a valid option. It should be noted that several results for the following analyses (especially for Pearson's r and R^2) would have been improved if the concentrations had been log transformed.

A few additional datasets are introduced in the coming chapters. Data descriptions of these datasets is undertaken using mean, SD and range, which are some of the most commonly used parameters for data description (Briggs et al. 2008; Gulliver & Briggs 2007; Boogaard et al. 2009; Pfeifer et al. 1999b; McNabola et al. 2009b; McNabola et al. 2009a; Praml & Schierl 2000). Datasets described in the following chapters show generally only a mild degree of skewness for their data distribution. Mean and SD may therefore be applied, instead of median and IQR applied above for highly positively skewed minute datasets.

3.7.2. Methods to Evaluate Model Performance

Several parameters are applied in this thesis, which measure the degree of similarity between two datasets. These parameters are foremost applied in this context to compare modelled and monitored particle concentrations, and as such to evaluate model performance. Two of the most commonly applied model performance parameters are Pearson's correlation, or Pearson's r , (r) and R^2 . Pearson's r measures the linear relationship of two variables. The range of r is from -1, a perfect negative linear correlation to +1, a perfect positive linear correlation. A value of 0 signifies the two variables are unrelated. R^2 , as applied here, is equal to the squared Pearson's r . The interpretation of

R^2 is therefore similar to the interpretation of r . All results of R^2 are however positive, ranging between 0 and 1 (Harris & Jarvis 2011).

<i>Coefficient of Divergence (COD)</i>	$COD = \left(\frac{1}{n} \sum_i \left(\frac{O_i - P_i}{O_i + P_i} \right)^2 \right)^{1/2}$
<i>Index of Agreement (IA)</i>	$IA = 1 - \frac{\sum_i (O_i - P_i)^2}{\sum_i (O_i - \bar{O} + P_i - \bar{O})^2}$
<i>Normalised Mean Square Error (NMSE)</i>	$NMSE = \frac{1}{n} \sum_i \frac{(P_i - O_i)^2}{\bar{P} \bar{O}}$
<i>Fractional Bias (FB)</i>	$FB = \frac{\bar{P} - \bar{O}}{0.5 (\bar{P} + \bar{O})}$

Table 27: Model Performance Parameters.

Several less commonly applied model performance parameters are presented with according equations in Table 27, with predicted values (P) and observed values (O) of samples (i). The coefficient of divergence (COD) measures the degree of similarity between values of two datasets. It has a range of between 0 and 1 with a COD value of 0 indicating the values are the same, a COD of 1 means maximum difference. Generally the interpretation is similar to Pearson's correlation (or R^2); however the COD provides a stricter metric. A COD of 0 means data values are identical, a Pearson's correlation of -1 or +1 on the other hand refers to a perfect linear correlation (Gaines Wilson & Zawar-reza 2006; Arku et al. 2008).

R^2 , Index of Agreement (IA or IOA), Normalised Mean Square Error (NMSE), and Fractional Bias (FB) are commonly applied together to describe model performance in the context of air pollution exposure modelling (Kousa et al. 2001; Beckx, Int Panis, Van De Vel, et al. 2009; Elbir et al. 2010; Gulliver & Briggs 2005). For this thesis this set of four parameters has as well been applied to describe and evaluate model performance.

R^2 has been introduced above and describes in this context the degree of linear correlation between modelled and observed. For the description of R^2 results the following broad categories are used: $0.0 > R^2 > 0.3$ = low, $0.3 > R^2 > 0.4$ = medium low, $0.4 > R^2 > 0.6$ = medium/moderate, $0.6 > R^2 > 0.8$ = medium high, $R^2 > 0.8$ = high.

The IA (sometimes also abbreviated IOA) measures the skill of the model to predict variations about the observed mean. IA values range between 0 and 1 with 0 indicating no agreement between variations and 1 indicating perfect agreement between modelled and observed. A value above 0.5 is considered a good model fit (Gaines Wilson & Zawar-reza 2006; Noth et al. 2011; Beckx, Int Panis,

Van De Vel, et al. 2009). The IA is sensitive to total differences between data values of observed and predicted, as well as to proportional changes.

The NMSE measures the distance between modelled and observed data points and as such is a measure of mean relative scatter, reflecting both systematic and unsystematic (random) errors. The NMSE parameter is normalised, so it is not dependent on unit. The NMSE is sensitive to occasional large errors due to the squaring process (Gaines Wilson & Zawar-reza 2006; Beckx, Int Panis, Van De Vel, et al. 2009).

The fractional bias (FB) between two datasets shows the degree of over- or underprediction of the predictions. The FB can range from -2 to +2, with -2 indicating extreme underprediction, and +2 indicating extreme overprediction; a FB of 0 indicates no over- or underprediction. FB values equal to -0.67 and +0.67 are equivalent to under- and over-predictions by a factor of two respectively (Beckx, Int Panis, Van De Vel, et al. 2009).

Applying the presented set of four parameters, the perfect model performance would be as follows: $R^2 = 1$, IA = 1, NMSE = 0, FB = 0. In the coming chapters model performance is evaluated using this set of four parameters presented above, first in chapter 4 for the evaluation of a temporally adjusted LUR model, developed as ambient base model, then in chapter 5 to compare performance of different indoor models applied from the literature, and finally in chapter 6 to evaluate performance of the personal exposure model.

3.8. Chapter Summary

In this chapter the different air pollution monitoring campaigns were introduced. The collected data was described, followed by the preparation of the data for analyses.

Overall, several main observations have been made for the collected data and can be summarised as follows:

- Monitored ambient concentrations were on average higher at roadside locations compared to background locations.
- Home outdoor concentrations showed only moderate short-term variability.
- In comparison to home outdoor concentrations, home indoor concentrations have higher short-term variability.
- Concentration variability indoors is related to the number of people actively occupying the home, as well as to cooking events.

- Times of cooking can result in extremely high concentrations and the recovery to pre-cooking concentrations can sometimes take several hours.
- Personal exposure variability follows a daily pattern related to times spent in different MEs.
- MEs differ both in terms of their typical median concentration, as well as in terms of their degree of variability.
- Simultaneously monitored walking and cycling exposure showed higher median concentrations for cycling compared to walking exposure. Simultaneously monitored cycling and in-bus exposure showed for $PM_{2.5}$ higher median concentration for the cycling, for PM_{10} the median was higher for in-bus exposure. Concentrations monitored at a background location for the times of transport monitoring were lower than exposure measured in any of the transport MEs

The described datasets form the basis for several analyses in the following chapters.

For chapter 4, the ambient monitoring campaign provides annual concentration averages at 21 locations in London and the Thames Valley region. These concentrations are used as part of the ESCAPE study for the development of LUR models.

In the first part of chapter 5 ratios for the adjustment of transport MEs are developed from ambient concentrations at the roadside and at background locations. These ratios are calculated from data collected during the transport monitoring campaign.

In the second half of chapter 5 models for home indoor MEs are compared. As a development to a commonly applied MB model, additional parameters for indoor source contributions are developed using data from the home indoor-outdoor campaign conducted at homes B, C, and D. Several different models for home indoor concentrations are then applied to estimate hourly concentrations at home A, for which concentrations were collected as part of the indoor-outdoor monitoring campaign. The performance of these models for the prediction of monitored indoor concentrations is then evaluated and compared.

In chapter 6 an integrated personal exposure model, the MEPEX model is built from components developed in chapters 4 and 5. Personal exposure predictions from the MEPEX model and other commonly used methods of exposure assessment are then applied to predict monitored concentrations collected by individual A during the personal exposure monitoring campaign. The performance of the different models is subsequently compared. Model transferability to other individuals is additionally tested for predictions of monitoring data collected for personal exposure by individuals B, C, and D.

4. Modelling Ambient Particle Concentrations

4.1. Introduction

One of the main components of the personal exposure model developed for this thesis is a model for ambient particle concentrations. In this chapter the development of a model for ambient air pollution is described. The modelling of ambient concentrations is of particular importance for the final personal exposure model as it is not only used to model the times spent at outdoor locations, but estimates are also used as input for models and adjustments of the other MEs.

Several models of ambient air pollution have been introduced in chapter 2. Few of these models however include both high temporal, as well as spatial resolution. For the application in a personal exposure model spatial variability is important to reflect exposure differences between addresses of a study population, as well as for locations visited when away from home. Temporal variability is as well of high importance, as personal exposure models are usually based on exposure variability at a less than daily level to reflect different MEs visited throughout the day. One of the most commonly applied model types for ambient air pollution models with high spatial and temporal resolution are dispersion models. Dispersion models work with the physical principle that pollution disperses away from a source, depending on meteorological conditions, building terrain etc. Model development of most particle dispersion models is complex and many studies rely on purchasing and applying existing modelling software. One major disadvantage of dispersion models is that data requirements for model applications are often high and in particular require detailed information on emissions sources in the study area. This type of data is however not always available.

An alternative to dispersion models are land-use regression (LUR) models. Compared to dispersion models data for LUR models (such as road geography and land use patterns) are often more routinely available and development of a model is in most cases less complex, allowing researchers to build models customised to their research area. LUR models are generally designed to predict a long-term average. Temporal adjustments have however been undertaken to predict shorter time periods by several studies (Aguilera et al. 2009; Aguilera et al. 2010; Brauer et al. 2008; Dons, Van Poppel, Int Panis, et al. 2014; Johnson et al. 2013; Mölter et al. 2012; Nethery et al. 2008; Slama et al. 2007; Pedersen et al. 2013).

The main objective of this chapter is to develop an alternative method to the more established dispersion model, which predicts ambient concentrations both with high spatial as well as high temporal accuracy, using a temporally adjusted LUR model. The applied LUR model has been developed as part of the ESCAPE study (contributions of the doctoral candidate are outlined below).

Temporal adjustments for the LUR model have been selected in a stepwise procedure, comparing performance of different adjustment methods for predictions at network monitoring stations. Performance of the final model was examined using model performance parameters.

While temporally adjusted LUR models have been applied in several studies little to no validation or model comparison has been undertaken to examine the performance of LUR models for short-term predictions, such as for days or hours. The comparison of LUR models to dispersion models for shorter time periods has additionally been pointed out as a possible focus for future research by Hoek et al. (2008). In order to evaluate the temporally adjusted LUR model developed in this chapter, model performance results for PM_{10} were evaluated in comparison with predictions by ADMS-urban, a dispersion model widely used by local authorities.

In following sections, first the ADMS-urban model is introduced. Then the development of a LUR model for the London area is described. Subsequently hourly temporal adjustments of the LUR estimates are developed in several steps. The performance of hourly LUR predictions is evaluated by predicting concentrations at network monitoring stations. The results for PM_{10} are subsequently compared with results from the ADMS-urban model.

4.2. Introduction and Application of the ADMS-urban Dispersion Model

4.2.1. Introduction

ADMS-urban is a well-established air pollution dispersion model used by many local authorities and other governmental bodies in the UK, as well as abroad. The model was developed by the Cambridge Environmental Research Consultants (CERC), who provide the ADMS-urban model as software, which can be purchased online (<http://www.cerc.co.uk/environmental-software/ADMS-Urban-model.html>). The focus of this section is to describe the set-up and application of the ADMS-urban software. ADMS-urban is applied to predict concentrations at London monitoring stations. The predictions are used later in the chapter to compare with results of the developed LUR model. ADMS-urban does not predict $PM_{2.5}$, which was therefore excluded from this comparison.

4.2.2. ADMS-urban Software Requirements

ADMS-urban calculates the dispersion of pollutants away from their sources. The dispersion equation is based on a quasi-Gaussian plume model nested within a simple trajectory model (Stocker et al. 2011). To calculate air pollution at a location it requires extensive input data on source locations and emissions, meteorological conditions, as well as several further input parameters:

1. Source data: depending on application; different sources can be added using sources summarised in a grid, point sources (e.g. stacks), road sources, or industrial source data.
2. Time-varying emissions: optional; adds temporal variability to the source input.
3. Meteorological data: dataset of wind speed, wind direction and one further input (e.g. cloud cover) as a minimum and optional information for example on temperature. Meteorological data may also convey time steps, e.g. day of year, which in turn determines the temporal resolution of the output.
4. Output points: ID, X, Y, and Z coordinates of predicted points
5. Terrain file: optional, to model effects of terrain on air pollution dispersion. The altitude of receptor points is included separately.
6. Background concentrations: background concentrations can be added as part of the software application or afterwards.

In addition to options regarding source input, levels can be set for Monin-Obukhov length and surface roughness. Both parameters are used to characterise conditions for air flow at the earth surface and influence the air dispersion calculation. Monin-Obukhov length is dependent on friction velocity and heat flux at the surface (e.g. influenced by urban heat island effect). Surface roughness is a measurement used to estimate smoothness of the earth surface (sea = smooth, wood or urban area = rough).

4.2.3. Application of the ADMS-urban Model

An overview over the input data as applied in this thesis is presented in Table 28. Source data for the London area (about 600km²) had already been prepared by colleagues for GEMS (Global and regional Earth-system (Atmosphere) Monitoring using Satellite and in-situ data), an EU 6th-framework project. For the GEMS project extensive source data had been prepared for the use in the ADMS-urban software. The source data had been acquired from the LAEI (London Atmospheric Emissions Inventory) and included about 60,000 road links of major roads with attached traffic data (vehicle number, composition, average speed), weighted by hour of day, as well as 1km grid squares, which included industrial and domestic sources, as well as small roads. The extensive data on major

roads was too large to be included into the ADMS-urban software directly. It was therefore cut into 51 geographical tiles and run separately. Each tile predicted the source contribution on air pollution for receptor points in the study area (regardless if they fell within the area of the tile). After repeatedly running the software for all tiles the results could simply be summed for each predicted location. Results from the separately run 1km grid square sources were then added to the particles calculated for major roads sources. Meteorological data was acquired from the London Heathrow station. Hourly data for wind speed, wind direction, cloud cover, and temperature for year 2010 were acquired from the met office. Heathrow station is located in an open area unobstructed by buildings or other obstacles and can therefore be considered a good indicator for meteorological conditions in the London area.

The altitude difference in London was considered negligible; therefore no terrain file was added. For Monin-Obukhov length and surface roughness the suggested levels given by CERC for major conurbations (>1 Million inhabitants) were applied, using a Monin-Obukhov length of 100 and a surface roughness of 1.5 (Cambridge Environmental Research Consultants Ltd. 2012).

Data Type	Description	Resolution	Source	Remarks
Road Sources	~63,000 individual road links with attributed traffic numbers, composition and speed	1:10,000	LAEI	Average hourly traffic changes added (average of 6 traffic counting stations in London, provided by King's College)
Grid Sources	Industrial, domestic, and small road sources	1km grid	LAEI	
Meteorological Data	Wind speed, wind direction, temperature, and cloud cover	Hour of the year	Met office	Heathrow station

Table 28: Input data for the ADMS-urban model

The predictions from the ADMS-urban model represent all source related concentrations of urban origin for locations in London. Regional background concentrations were added in a next step. Regional background concentrations are generally similar over a relatively large area and change slowly over time with major changes usually not occurring within less than a few hours (Jönsson et al. 2013; World Health Organisation Europe 2006b), adjusting for these particles can therefore be undertaken using a comparably simple method. In order to adjust ADMS model predictions for background concentrations many studies have added a spatially indifferent time-series of concentrations (Hänninen et al. 2009; Fecht 2011; Gulliver & Briggs 2005; Blangiardo et al. 2011; Tonne et al. 2008). The most direct way to add a time-series of regional background is by using concentrations from rural background sites or their average. More complex methods were sometimes applied when monitored concentrations from rural background sites were not available (Gulliver & Briggs 2005; Tonne et al. 2008).

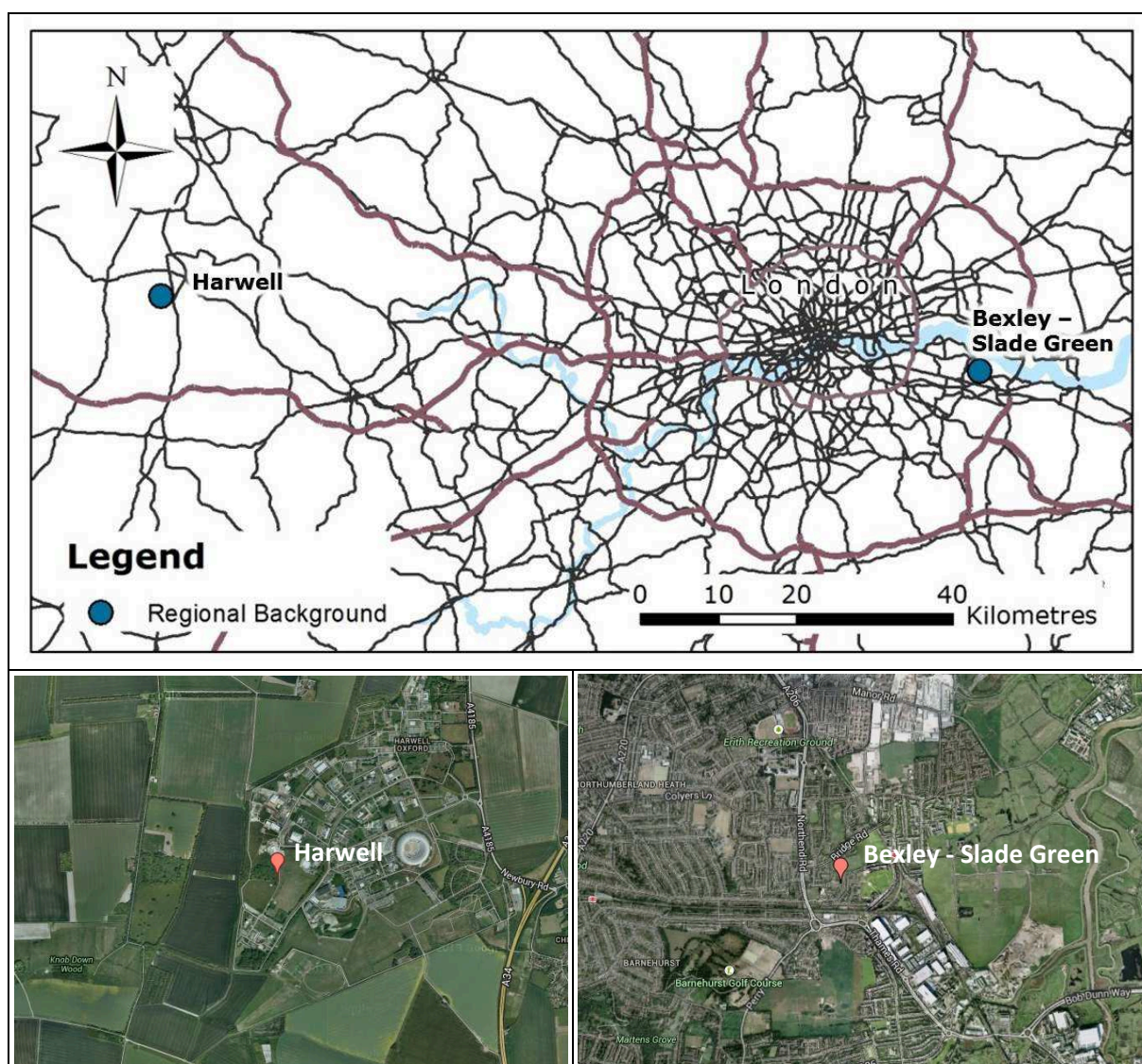


Figure 26: Site locations of monitoring stations used to adjust for regional background concentrations: Harwell (left) and Bexley – Slade Green (right). Road data source, top: Meridian. Source, bottom: Google maps (<https://maps.google.co.uk>, accessed: 17th March 2014)

The AURN network provided data for air pollution monitoring stations in Britain online (<http://uk-air.defra.gov.uk/networks/network-info?view=aurn>). In southwest England only one rural background site, Harwell (about 75 km west of Central London) provided PM_{10} concentrations for 2010 within a 100km radius around London (see Figure 26). A decision was made to include Bexley - Slade Green, a London suburban background site (Figure 26), as a regional background average with Harwell to get a more balanced estimate and to provide data points for missing data at Harwell station. Bexley was selected for this purpose for several reasons: 1). It is located east of central London and provides to some degree a geographical balance to Harwell, which is located West of

London. 2) The site is located at the border of London suburban living quarters stretching to the West, and open countryside to the East (see Figure 26) with considerable areas of open landscape close to the site. 3) The station is one of the London monitoring network stations furthest away from the centre, therefore being subject to relatively little influence from high source activity in central London. 4) The wider area around the site shows no major sources: no industrial sites were detected within 1km around the station (using Google Streetview), and the nearest major road is about 250m away.

To decide on the temporal precision of the regional background data, several facts had to be considered. The ADMS-urban model was applied to predict hourly concentrations; temporal periods for regional background should not be much longer to match the high temporal precision. On the other hand, changes of regional background concentrations are mainly driven by large air masses, major concentration changes therefore happen slowly (less than hourly). Additionally, the regional background stations are relatively far away from central London (Harwell station is more about 70km from Central London, Bexley - Slade Green is located inside the M25, but still about 23km from Piccadilly Circus), it is unrealistic to assume hourly precision over such a distance. Several studies have applied daily concentration averages (Briggs 2007; Blangiardo et al. 2011). A slightly shorter period of bi-daily concentrations was preferred here, as this matches the hourly ADMS-urban predictions better. In addition, there is some evidence that a difference between night-time and daytime pollution concentrations exists at a regional level (Beckx, Int Panis, Van De Vel, et al. 2009; Bayer-Oglesby et al. 2004). Night-time was defined as 8pm to 6am representing approximate hours of darkness during equinoxes, as well as hours of traffic below the daily average (Department for Transport 2011) indicative of low human activity at night. The bi-daily regional averages were added to the modelled ADMS-urban concentrations. Five bi-daily steps had missing data for the regional average, resulting in 61 missing data points for the hourly ADMS-urban predictions of 2010.

4.3. ESCAPE LUR Model Preparation and Development

4.3.1. Introduction

As described above the ADMS-urban model requires relatively large data input and potentially long processing times. A LUR model is presented here as an alternative option for short-term modelling of particles. First, the development of the LUR model for the London and the Thames Valley region as part of ESCAPE (European Study of Cohorts for Air Pollution Effects), an EU 7th framework project is described. Brief introductions to the project have been given in chapters 2 and 3. One of the main

objectives of the ESCAPE study was “to develop a flexible methodology for assessment of long-term population exposure to air pollution...” (<http://www.escapeproject.eu>). LUR models for PM_{2.5} and PM₁₀ for all ESCAPE study areas, including London and the surrounding Thames Valley area were developed by the project. The doctoral candidate worked as part of the ESCAPE project on the development of the London & Thames Valley model (her involvement is detailed below).

As a further development beyond the scope of the ESCAPE project, temporal adjustments of the LUR model using monitoring data were developed as part of this thesis. In a stepwise process different ways to apply temporal adjustments to the LUR model were examined and compared. Furthermore, model performance of the final model is examined in comparison to predictions by the ADMS-urban model (PM₁₀ only). The short-term LUR model thus developed is used in further chapters as part of the personal exposure model.

4.3.2. Data Preparation and Model Development

Within the ESCAPE project a standardised protocol was applied for LUR model development, which is described in detail in the study manual (Beelen & Hoek 2010) and is outlined below. LUR models for the prediction of particulates within the London & Thames Valley region were developed using adjusted annual average concentrations of PM from a bespoke monitoring campaign described in chapter 3. In a first step the data sets presented in Table 29 were prepared and integrated into a GIS, and potential explanatory variables were derived for the locations of the monitoring sites. This for example included the sum of road length in buffers of different sizes (Radius: 25m, 50m, 100m, 300m, 500m, and 1000m) around each location, or altitude at the sites. Buffers can be calculated in a GIS as circular areas around a site. The calculation of different buffer sizes for each type of variable was undertaken to evaluate the impact of predictor variables at different spatial scales. The selection of minimum and maximum buffer sizes for LUR input variables was based on known dispersion patterns. The effect of traffic source emissions for example reduces rapidly with distance from road, which is for example reflected in small buffer sizes of 25m or 50m for traffic variables. Larger buffer sizes around roads, such as 1000m were included to reflect the sum of emissions from roads in the area. Land use buffers were generally chosen larger than road variable buffer sizes (from 100 to 5000), as they do not reflect a specific source, but represent diffuse sources over an area. 30 different principle predictor variables (not counting different variations or buffer sizes of the same variable) were produced, which are shown in Table 30.

Data Type	Name	Description	Resolution	Version	Source	Remarks
Land Use Data	CORINE 2000	Vector data	1:100,000	2000	European Environmental Agency	44 land-use categories grouped into 6 classes: high and low density residential land, industry, ports, urban green and natural land
Roads	Meridian 2	Vector data	1m	2009	Ordnance Survey	
Traffic Intensity	National Traffic Estimates	Traffic counts & grid reference for main roads	-	2009	Department for Transport, Road Traffic Statistics	Data was attached to Meridian 2 dataset
Population Density	Headcount Population Data	Population estimates based on 2001 census	Postcode level (points)	2001	Office of National Statistics	-
Altitude	Digital elevation data (SRTM)	Raster data	90m	-	CGIAR-CSI GeoPortal	-

Table 29: Datasets used to calculate ESCAPE-LUR model predictor variables for the London and the Thames Valley study area. Source: Eeftens et al. (2012), modified. SRTM = Shuttle Radar Topography Mission

The LUR model was developed using standard linear regression. In a forward stepwise approach each candidate predictor variable was introduced individually as a potential variable to enter in the model. The resulting R^2 were compared and the variable with the strongest predictive power (R^2) was chosen to form part of the final regression equation. In the next step all other candidate predictor variables were again introduced individually and the next variable that raised the R^2 highest was chosen to remain in the model. This process was repeated until no added variable could raise the predictive power (R^2) of the model further. The introduction of variables was closely monitored and several rules applied (Beelen & Hoek 2010):

1. A variable needed to improve the R^2 by at least 1% to enter into the equation.
2. Minimum significance of all entered variables was 0.05.
3. Each variable had a predefined direction of effect (Table 30): e.g. increased nearby traffic intensity was expected to have a positive effect on the slope, as it should increase concentrations; urban green was expected to have a negative effect as it would provide space for particle dispersion.
4. Effects of 1, 2 and 3 should not change for any included variable if another variable was added.

Table 30: Principle predictor variables for the ESCAPE-LUR model with predefined variable names, units, defined buffer sizes, and directions of effect. Variables chosen for London and the Thames Valley models (PM_{2.5} and PM₁₀) are highlighted in blue and bold. Source: Eeftens et al. (2012).

GIS dataset	Predictor variable	Name variable ¹	Unit	Buffer size (radius of buffer in meter)	Direction of effect
Background					
-	Coordinate variables ⁴	XCOORD, YCOORD	m	NA	NA
CORINE	Surface area of high density residential land	HDRES_X	m ²	100, 300, 500, 1000, 5000	+
CORINE	Surface area of low density residential land	LDRES_X	m ²	100, 300, 500, 1000, 5000	+
CORINE	Surface area of low and high density residential land combined	HLDRES_X	m²	100, 300 , 500, 1000, 5000	+
CORINE	Surface area of industry	INDUSTRY_X	m ²	100, 300, 500, 1000, 5000	+
CORINE	Surface area of port	PORT_X	m ²	100, 300, 500, 1000, 5000	+
CORINE	Surface area of urban green ²	URBGREEN_X	m ²	100, 300, 500, 1000, 5000	-
CORINE	Surface area of semi-natural and forested areas ³	NATURAL_X	m ²	100, 300, 500, 1000, 5000	-
CORINE	Sum of Urban green and Semi-natural and forested areas	GREEN_X	m ²	100, 300, 500, 1000, 5000	-
CORINE	Surface Area of Water	WATER_X	m ²	100, 300, 500, 1000, 5000	-
Population density	Number of inhabitants	POP_X	N(umber)	100, 300, 500, 1000, 5000	+
Household density	Number of households	HHOLD_X	N(umber)	100, 300, 500, 1000, 5000	+
Altitude	Altitude	SQRALT	m	NA	-
Traffic⁶					
Local road network	Traffic intensity on nearest road	TRAFNEAR	Veh.day ⁻¹	NA	+
Local road network	Inverse distance and inverse squared distance to the nearest road	DISTINVNEAR1 DISTINVNEAR2	m ⁻¹ , m ⁻²	NA	+
Local road network	Product of traffic intensity on nearest road and inverse of distance to the nearest road and distance squared	INTINVDIST INTINVDIST2	Veh.day ⁻¹ m ⁻¹ Veh.day ⁻¹ m ⁻²	NA	+
Local road network	Traffic intensity on nearest major road ⁶	TRAFMAJOR	Veh.day ⁻¹	NA	+
Local road network	Inverse distance and inverse squared distance to the nearest major road⁶	DISTINVMAJOR1 DISTINVMAJOR2	m⁻¹, m⁻²	NA	+
Local road network	Product of traffic intensity on nearest major road and inverse of distance to the nearest major road and distance squared⁶	INTMAJORINVDIST INTMAJORINVDIST2	Veh.day⁻¹m⁻¹ Veh.day ⁻¹ m ⁻²	NA	+

Local road network	Total traffic load of major roads in a buffer (sum of (traffic intensity * length of all segments)) ⁶	TRAFMAJORLOAD_X	Veh.day ⁻¹ m	25, 50, 100, 300, 500, 1000	+
Local road network	Total traffic load of all roads in a buffer (sum of (traffic intensity * length of all segments))	TRAFLOAD_X	Veh.day ⁻¹ m	25, 50, 100, 300, 500, 1000	+
Local road network	Heavy-duty traffic intensity on nearest road	HEAVYTRAFNEAR*	Veh.day ⁻¹	NA	+
Local road network	Product of Heavy-duty traffic intensity on nearest road and inverse of distance to the nearest road and distance squared	HEAVYINTINVDIST HEAVYINTINVDIST2	Veh.day ⁻¹ m ⁻¹ Veh.day ⁻¹ m ⁻²	NA	+
Local road network	Heavy-duty traffic intensity on nearest major road⁶	HEAVYTRAFMAJOR	Veh.day⁻¹	NA	+
Local road network	Total heavy-duty traffic load of major roads in a buffer (sum of (heavy-duty traffic intensity * length of all segments)) ⁶	HEAVYTRAFMAJORLOAD_X	Veh.day ⁻¹ m	25, 50, 100, 300, 500, 1000	+
Local road network	Total heavy-duty traffic load of all roads in a buffer (sum of (heavy-duty traffic intensity * length of all segments))	HEAVYTRAFLOAD_X	Veh.day ⁻¹ m	25, 50, 100, 300, 500, 1000	+
Central road network	Road length of all roads in a buffer	ROADLENGTH_X	m	25, 50, 100, 300, 500 , 1000	+
Central road network	Road length of major roads in a buffer ⁵	MAJORROADLENGTH_X	m	25, 50, 100, 300, 500, 1000	+
Central road network	Distance to the nearest road	DISTINVNEARC1 DISTINVNEARC2	m ⁻¹ , m ⁻²	NA	+
Central road network	Inverse distance and inverse squared distance to the nearest major road ⁵	DISTINVMAJORC1 DISTINVMAJORC2	m ⁻¹ , m ⁻²	NA	+

¹ Variable name: Combining name and buffer size, e.g. for HDRES_X: HDRES_100, HDRES_300, HDRES_500, HDRES_1000, HDRES_5000

² CORINE Urban green is the sum of CORINE classes 141 and 142

³ CORINE semi-natural is the sum of CORINE classes 311, 312, 313, 321, 322, 323, 324, 331, 332, 333, 334, 335, 411, 412, 421, 422, 423, 512, 521, 522 and 523

⁴ Variables were only offered if a model has been developed to test if the model with more explicit variables could be improved with these variables (describing slow trends in background).

⁵ Definition of major road for central road network: classes 0, 1, and 2 (+ classes 3 and 4 based on local knowledge and decision)

⁶ Definition of major road for local road network: road with traffic intensity > 5,000 mvh/24h

NA signifies not applicable

The concentrations collected during each seasonal monitoring campaign (described in chapter 3) were weighted to the annual average for the analysis as follows: 2-weekly average concentrations measured at the reference site, which operated during the whole monitoring period, were used to calculate the ratio to the reference site's annual mean. These ratios were subsequently applied as adjustment to all sites for each round (Eeftens, et al., 2012). Several tests and checks were applied to the final model to assure the assumptions underlying the regression analyses were met:

1. The selected regression variables were checked for colinearity. Regression results with multiple variables can have correlated explanatory variables, which increase the R^2 adj. without explaining the dependent variable. The Variance Inflation Factor (VIF) calculates the colinearity between variables. Variables with a VIF of below 5 are generally considered to have low colinearity (Harris & Jarvis 2011).
2. Results in a regression can be influenced by single observations in the dataset. In order to check for unusually influential data points Cook's D was calculated. Cook's D (or Cook's distance) is a measure of each observation's influence on the regression line. Values with a Cook's D above 1 should be further investigated (Harris & Jarvis 2011).
3. Normal distribution of the residuals was ensured by looking at the residual distribution in histograms.
4. Spatial data has the tendency that locations near to each other can be more similar. This tendency, called spatial autocorrelation can violate the assumption of independent data distribution in a regression. Spatial autocorrelation was tested using Moran's I statistics (Eeftens et al. 2012; ESCAPE project 2010). A Moran's I close to zero signifies low spatial autocorrelation. Moran's I values can be transformed to z scores for which values greater than 1.96 or smaller than -1.96 signify high autocorrelation significant at the 5% level (Harris & Jarvis 2011).

Model validation was performed by leave-one-out cross-validation (LOOCV). For LOOCV the predictor variables are fixed and the regression is run for $n-1$ (in this analysis for $20-1=19$ sites), leaving each one of the data points out in $n-1$ repeated regression analyses. The resulting R^2 adj. is then compared with the R^2 adj. of the full dataset (Wang et al. 2013).

4.3.3. ESCAPE-LUR Models for London and the Thames Valley

By applying the supervised stepwise regression procedure described above, two explanatory variables were selected for the $PM_{2.5}$ LUR equation. The first included variable was INTMAJORINVDIST, defined as the "Product of traffic intensity on nearest major road and inverse of

distance to the nearest major road” using the unit of $\text{veh.day}^{-1}\text{m}^{-1}$. The second variable was ROADLENGTH500, defined as “Road length of all roads in a buffer of 500m” with metres as units.

Three explanatory variables were selected for PM_{10} . The first variable was DISTINVMAJORC1, which is defined as the “Inverse Distance to the nearest major road” in m^{-1} . The second variable was HEAVYTRAFMAJOR, defined as “Heavy-duty traffic intensity on nearest major road” in veh.day^{-1} . The third variable was HLDRES300, defined as the “Surface area of low and high density residential land combined within a 300m radius” in m^2 .

Variable	Size fraction	Measurement Units	10 th Percentile	Median	90 th Percentile
INTMAJORINVDIST	$\text{PM}_{2.5}$	(veh/day)/m	20	122	1770
ROADLENGTH_500	$\text{PM}_{2.5}$	m	7359	11258	15066
HEAVYTRAFMAJOR	PM_{10}	veh/day	450	1157	3439
DISTINVMAJORC1	PM_{10}	1/m	0.001	0.0035	0.087
HLDRES_300	PM_{10}	m^2	190188	281987	282618

Table 31: Measurement units and basic statistics of selected explanatory variables for the ESCAPE-LUR model of London and the Thames Valley. veh = vehicles, Source: Eeftens et al. (2012).

Table 31 provides an overview of the different units and summary statistics of the selected explanatory variables, the percentiles and median presented in the table highlight the differences in the range of these variables from a median of 0.0035 for DISTINVMAJORC1 to a median of 281987 for HLDRES_300. Final results of the regression analyses are presented in Table 32. Adjusted R^2 are high with 0.804 for $\text{PM}_{2.5}$ and 0.900 for PM_{10} . SEE are low both for $\text{PM}_{2.5}$ with 1.29 and for PM_{10} with 1.48. The Significance of the constant (b_0) and different coefficients are all below 0.05 and the co-linearity of the different variables in the regression is low with VIF below 2.

	R^2 adj.	SEE	b_0			b_1			b_2			b_3		
			b	Sig		b	Sig	VIF	b	Sig	VIF	b	Sig	VIF
$\text{PM}_{2.5}$	0.804	1.29	7.194	0.000		0.001381	0.000	1.696	0.000265	0.017	1.696	-	-	-
PM_{10}	0.900	1.48	11.401	0.000		76.990	0.000	1.108	0.001353	0.000	1.186	0.000013	0.030	1.079

Table 32: Regression results for the ESCAPE-LUR model of London and the Thames Valley of $\text{PM}_{2.5}$ and PM_{10} . Variables underlying coefficients: For $\text{PM}_{2.5}$ b_1 = INTMAJORINVDIST, b_2 = ROADLENGTH500; for PM_{10} b_1 = DISTINVMAJORC1, b_2 = HEAVYTRAFMAJOR, b_3 = HLDRES300; Sig = significance; VIF= Variance Inflation Factor; SEE = Standard Error of the Estimate

Results for Cook’s D, Moran’s I and the LOOCV validation are presented in Table 33. Cook’s D remained well below 1 for both particle size fractions, indicating that there is no single influential

data point. Moran's I is close to 0 for both particle size fractions and z values are between -1.96 and 1.96, which signifies low spatial autocorrelation. LOOCV results show adjusted R^2 slightly lower than the original LUR regression with 0.758 for $PM_{2.5}$ and 0.874 for PM_{10} , which indicate a small over-estimation of the predictive power in the originally developed regression equation.

	Cook's D	Moran's I		LOOCV Validation	
		Moran's I	z value	R^2 adj.	SEE
$PM_{2.5}$	0.425	-0.19	-1.26	0.758	1.44
PM_{10}	0.162	-0.13	-0.80	0.874	1.52

Table 33: Tests and validation results for the ESCAPE-LUR model of London and the Thames Valley for $PM_{2.5}$ and PM_{10} .
SEE = Standard Error of the Estimate

The final annual LUR models for London & the Thames Valley area are as follows:

Equation 4

$$LUR_{PM_{2.5}}: 7.194 + 0.001381 * INTMAJORINVDIST + 0.000265 * ROADLENGTH500.$$

Equation 5

$$LUR_{PM_{10}}: 11.401 + 76.990 * DISTINVMAJORC1 + 0.001353 * HEAVYTRAFMAJOR + 0.000013 * HLDRES300.$$

Equation 4 shows the regression formula as developed for $PM_{2.5}$ ($LUR_{PM_{2.5}}$) and Equation 5 shows the regression formula for PM_{10} (Equation 5), both using the variables as described above. Results presented in this section have shown that both the LUR model for $PM_{2.5}$ (Equation 4) as well as for PM_{10} (Equation 5) have been developed with high predictive power. The stability of the models has additionally been tested in a validation. It should be noted however, that additional evaluation of the ESCAPE models described by Wang et al. (2013) has shown that the small number of data points to some extent inflated R^2 results for the model and for the LOOCV. Results have however been found to still predict a substantial fraction of the variation in the monitored data. Following these results, the developed LUR equations have consequently been used for this thesis to make long-term predictions at locations in London.

NB: Data preparation and calculation of variables, as well as development of LUR models with preliminary monitoring data formed part of this thesis. The regression equation and validation developed with the final monitoring data (presented as results above) was however undertaken by colleagues within the ESCAPE project. The contribution of the doctoral candidate to the ESCAPE study

are to some extent reflected in the following papers: Wang et al. (2013) and Eeftens et al. (2012) for which the doctoral candidate is co-author. Additional related work on developing LUR models as part of the ESCAPE study was undertaken by the doctoral candidate for particle composition, as well as for NO, and NO_x, see also: Cyrus et al. (2012) and De Hoogh et al. (2013), for which the doctoral candidate is as well co-author.

4.4. Incorporating Temporal Variability in the LUR Model

4.4.1. Introduction

In this section a method for temporal adjustment of LUR predictions to hourly estimates is described. Several existing studies on air pollution exposure have used temporally adjusted LUR models (see Table 34). Adjustments in these studies were undertaken using concentrations measured at routine monitoring stations. The studies followed two different approaches, 1) by applying adjustments directly using monitored concentrations for a certain time period, e.g. days, divided by the long-time (e.g. annual) average, and 2) by temporal pattern, such as long-term average for each hour of the day.

Study	No. of monitoring sites	Pollutant	Temporal period	Method
Aguilera et al. (2009;2010)	1	NO ₂	Days, then averaged to individual pregnancy trimesters	Direct
Brauer et al. (2008)	1	NO, NO ₂ , PM _{2.5} , black carbon	Months	Direct
Dons, Van Poppel, Int Panis, et al. (2014); Johnson et al. (2013)	1	black carbon, NO ₂	Days	Direct
Mölter et al. (2012)	5	NO ₂	Hours	Monthly Direct & hourly pattern
Nethery et al. (2008)	11 (NO/NO ₂), 6 (PM _{2.5}); 3 nearest each location	NO, NO ₂ , PM _{2.2}	Approach1: months	Temporal pattern
			Approach 2: hours, then averaged to 48 hours for analysis	Direct
Pedersen et al. (2013)	1	NO ₂ , PM _{2.5} , PM ₁₀	Days, then averaged to individual pregnancy trimesters	Direct
Slama et al. (2007)	1	NO ₂ , PM _{2.5}	Days, then averaged to individual pregnancy trimesters	Direct

Table 34: Studies which applied temporal adjustments to LUR model estimates.

Direct adjustments have been undertaken by Brauer et al. (2008) predicting monthly exposure both of particles (black carbon, PM_{2.5}) and of Nitrogen Oxides (NO, NO₂) for periods of pregnancies.

Johnson et al. (2013) use direct daily adjustments to predict black carbon and NO₂. The model by Johnson et al. (2013) was then used by Dons, Van Poppel, Int Panis, et al. (2014) to predict exposure of school children. Studies by Aguilera et al. (2009, 2010) have also used direct LUR adjustments to predict daily NO₂ concentrations, which were then averaged for individual pregnancy trimesters of a birth cohort. Two other studies have applied the same approach to calculate individual pregnancy trimesters. One of these studies is by Slama et al. (2007) studying birth weights for just over 1,000 births of a cohort in Munich, Germany. The other study is by Pedersen et al. (2013). This study pooled data from 14 birth cohorts in 12 European countries to study the relationship of air pollutants to birthweight as part of the ESCAPE project. Another study on exposure during pregnancies by Nethery et al. (2008) used direct hourly adjustments in one of their approaches, these were then aggregated to 48 hour averages to match periods of personal monitoring. Adjustment by temporal pattern was used by Nethery et al. (2008) for his second approach for predictions of month of the year, applying adjustments for monthly averages of six years. Mölter et al. (2012) used a mixed approach, first adjusting by a monthly ratio (for the same year), and then using an hour-of-day pattern to predict hourly NO₂ concentrations for school children.

The number of monitoring stations and how they were applied also differs between the mentioned studies. Several studies used one site for the whole study area (Aguilera et al. 2009; Aguilera et al. 2010; Brauer et al. 2008; Dons, Van Poppel, Int Panis, et al. 2014; Slama et al. 2007; Pedersen et al. 2013), Mölter et al. (2010a, 2012) however used the average of five stations within the area. A different approach was undertaken by Nethery et al. (2008), who used weighted concentrations from the nearest three stations to adjust each LUR prediction point (approach 2). This approach is based on the geographical premise that closer locations tend to be more similar, called spatial autocorrelation (Harris & Jarvis 2011). Monitoring stations close to a location should accordingly predict better than stations further away. It is however not known to what extent this would be reflected for stations in Central London, a relatively small study area with high local variability in PM concentrations.

All studies mentioned here use an adjusted LUR model to predict exposure in an epidemiological context. Comparisons between short-term LUR predictions and monitored concentrations have only been undertaken using concentrations from personal exposure monitoring. Monitored personal exposure however is not only dependent on ambient concentrations, but also highly depends on ME context. It is therefore difficult to draw conclusions about the model performance from these studies.

The review of studies, which temporally adjusted LUR predictions lead to several questions about this approach:

- Does the nearest monitoring station or the average of monitoring stations in the area better reflect the temporal variability at a location? Which of these versions should be used for calculating adjustments for LUR models?
- Which approach works better: Directly using variability monitored at network stations or using repeated temporal pattern of concentration variability (such as hour of day or day of week) for predictions?
- To what extent are either of these approaches transferable in time?
- To what extent are either of these approaches transferable in space?
- How well does a temporally adjusted LUR model perform in comparison to other models, in particular to dispersion models?

To answer these questions a systematic approach was applied to develop and test temporal adjustments for LUR predictions step by step. Step 1 to step 3 develop and test different types of temporal adjustments. In step 4 the adjustment which showed best agreement with the concentration variability at different locations is applied and its performance is evaluated in more detail:

- Step 1: Evaluating if 1) the nearest background station or 2) an average of background stations better reflects variability at a location in London using Pearson's correlation. The approach which better reflected concentration variability was selected to be applied in further steps.
- Step 2: Developing two versions of temporal adjustments (calculated from 2010 concentrations of a set of London monitoring stations): Adjustment A) direct application of monitored concentrations; Adjustment B) using temporal pattern to predict short-term concentrations.
- Step 3: Comparing predictions of monitored concentration variability between temporal adjustments A and B
 - a) Comparing LUR adjustments A and B for predictions of 2010 concentrations at locations in London, using Pearson's correlation.
 - b) Testing temporal transferability by applying temporal adjustments calculated from 2010 concentrations to predict concentrations in 2011 at locations in London, using Pearson's correlation.
 - c) Testing spatial transferability of adjustments A and B by predicting monitoring stations outside London using Pearson's correlation.
 - d) Based on results for 2010 predictions, as well as for transferability, one of the two adjustment methods (A or B) was selected for further application in step four.

- Step 4: Evaluating model performance of temporally adjusted LUR concentrations (as developed in steps 1, 2, and 3) for locations in London by:
 - Looking at performance parameters for predictions of monitoring stations in London,
 - Comparing performance with predictions calculated with the ADMS-urban model (PM₁₀ only).

For the calculation of temporal adjustments, as well as for the predicted locations, network monitoring stations in London are used, which are described below. The outlined evaluation and development of a temporally adjusted LUR model is undertaken for hourly, daily and weekly concentrations. Hourly ambient predictions underlie calculations of most personal exposure models, including the model developed for this thesis. A need for the evaluation of temporally adjusted LUR models beyond the applications in this thesis was however recognised. Daily and weekly averages of particle concentrations are commonly used in short term exposure studies (see chapter 2) and are therefore added to this analysis.

A spatial focus on London (see details below) was taken in this analysis for three reasons: 1) London has a dense network of monitoring stations with freely available data, unmatched by any other area in the UK, which could be used for LUR predictions in this analysis, 2) The ESCAPE-LUR model was developed for London and the Thames Valley with most monitors located within London, and 3) London is the focus for applications of the personal exposure model developed in this thesis. The question of spatial transferability was however addressed by testing correlations with temporal variability at a selection of locations outside London. In the following sections, first the data used from the network monitoring stations are described, then steps one, two, three, and four are presented, followed by a discussion.

2010 was used as time period for this application, as the ESCAPE-LUR model predicts 2010 concentrations, and most personal exposure modelling in this thesis was undertaken for this time period. It was however tested if the temporal adjustments would be transferable to another time period by applying adjustments based on 2010 data to particle concentrations measured at monitoring stations in 2011.

4.4.2. Network Monitoring Stations and Data

In preparation for the following analysis long-term LUR estimates for PM_{2.5} and PM₁₀ were calculated using the ESCAPE-LUR model for London & Thames Valley, as described above. Monitoring data from the London Air Quality Network (LAQN) was used both to calculate short-term LUR adjustments and

to provide a test dataset for predictions. Hourly ratified concentrations for monitoring stations in the London area are provided online by the LAQN (<http://www.londonair.org.uk/>). Monitoring network station data for locations outside London was acquired from the Department of Environment Food and Rural Affairs (DEFRA) online (http://uk-air.defra.gov.uk/data/data_selector). Only stations with data availability of above 75% were included.

For the analysis a set of network monitoring stations (or sites) was used to calculate adjustments to the LUR model (development sites). For these development sites it was important that the concentrations should broadly reflect pollution representative for personal exposure, as the model for ambient concentrations was intended to underlie exposure predictions. Background locations were generally considered most suitable as development sites. Background monitoring stations generally provide a more representative picture for a whole area and are less influenced by their specific location compared to e.g. roadside or industrial locations (<http://uk-air.defra.gov.uk/networks/site-types>, accessed: 14th April 2014). Identification of background locations can be undertaken either by using the existing site type definitions for each network monitoring station (such as urban background, or roadside) or by using distance from major road (Vineis et al. 2007). Using distance from major road can provide a consistent and reproducible method, especially useful for large study areas with many sites. Using distance from major road however relies on the assumption that the particles can distribute freely and continuously away from the source. In highly built-up areas, such as London, this is however often not the case. Major roads in major cities are frequently framed by tall buildings, largely obstructing free propagation of particles. The site type definitions by the LAQN were therefore preferred for the study area of London. The LAQN definitions are generally based on distance to major road, but takes local characteristics into account.

Network monitoring sites in London are categorised by the LAQN into four site types: Roadside, urban background, industrial, and suburban; of which roadside stations are the most numerous, followed by urban background stations (<http://uk-air.defra.gov.uk/networks/site-types>, accessed: 14th April 2014; <http://www.londonair.org.uk/london/asp/publicdetails.asp?region=0>, accessed: 14th April 2014):

1. Roadside stations are located in the direct vicinity to major roads and concentrations at the site are influenced by localised emissions from traffic on these roads.
2. Urban background sites are located within a built-up area with houses, streets etc. (this can include areas outside a city), but away from or separated from the direct influence of sources.
3. Industrial sites are located so that the site is influenced by industrial sources nearby.

4. Suburban sites are (according to the definition by the DEFRA) located between built-up areas and open landscape (fields, wetlands etc.). “Suburban” in this context does not mean suburban living quarters or suburbia and for London includes for example a site in the London Wetland Centre at Richmond upon Thames or a site (Bexley Thamesmead) close to the Thames River and the Thamesmead Ecology Study Area.

For the development sites a decision was made to use urban background stations according to the LAQN definition. Urban background stations were considered to best reflect concentrations for areas where people spent most of their time (i.e. home and work locations).

A second set of network monitoring stations, apart from the development sites, was used to provide locations for predictions (prediction sites). There was no requirement for these stations to be directly representative of personal exposure, as the focus was to evaluate concentration predictions for a generic set of locations. For the prediction sites, roadside monitoring stations were used, as they are by far the most numerous site type in London and had not been included in the development sites. Both for step 1, as well as for step 4, analyses were however in addition undertaken for development sites, as well as for prediction sites, for the following reasons: For step 1 (nearest monitoring station versus average of monitoring stations) it was tested if the background locations (selected for development sites) would show higher correlations with other background locations in contrast to roadside locations (selected for prediction sites). In step 4, predictions of the final model were as well undertaken for development sites to evaluate the final model performance not only for roadside locations, but also for background locations. In order to predict at development sites when using the average of several background monitoring stations the LOOCV (leave-one-out cross-validation) approach was applied by predicting one development site with the average of the other development sites. Altogether 13 monitoring stations for $PM_{2.5}$ (4 background, 9 roadside) and 36 monitoring stations for PM_{10} (10 background, 26 roadside) were selected.

For PM_{10} a large number of monitoring sites was available for the area. A geographical limit for site inclusion was set at the North and South Circular Road around central London (see Figure 28). For $PM_{2.5}$ less sites were available, monitoring sites from a wider geographical area were therefore included, setting a limit at the M25 (see Figure 27). The names for each station and their ID are presented in the tables at the bottom of the two figures together with number of data points, mean, standard deviation and range of hourly concentrations. Basic statistics of daily concentrations and weekly concentrations are added in Appendix H.

The data as downloaded from the LAQN websites did not require further cleaning, as it had already gone through a careful four stage ratification process including automatic and manual checks of

unusual values. The concentrations were therefore considered to generally reflect pollution concentrations well and further removal of extreme values was tried to be avoided. PM₁₀ station number 19 (Newham Cam Rd) however showed an extreme range of hourly concentrations of 2999µg/m³ (see Figure 28). The high concentrations in the dataset considerably changed results for this station. For station 19 therefore 10 outliers were removed according to the described removal of outliers in chapter 3. Both results for the original data and after removal of outliers (station ID: 19b) are presented in all tables and graphs. In the text results are however summarised referring to station 19b, without outliers.

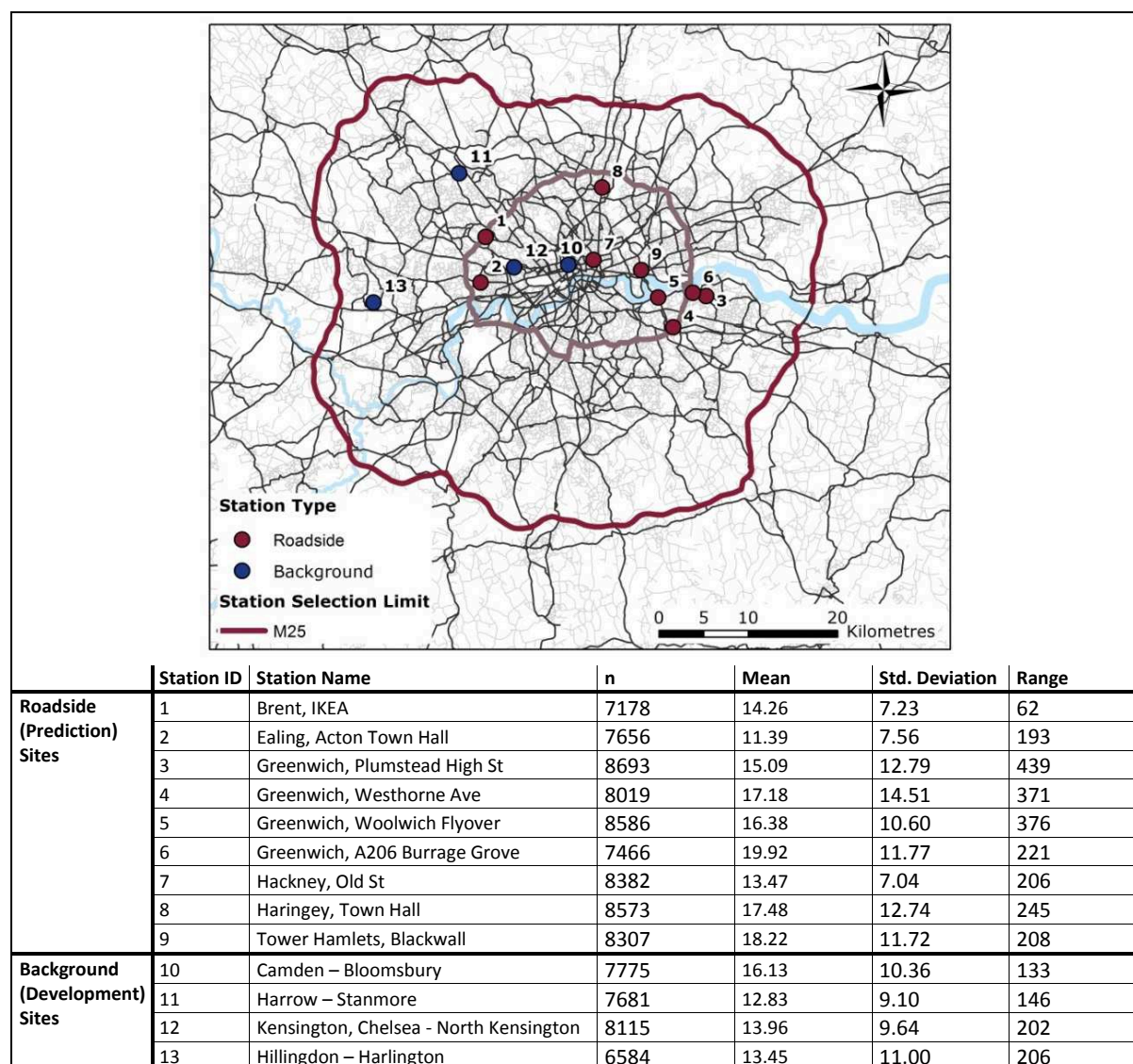


Figure 27: Locations of PM_{2.5} monitoring stations in London and limit for station selection (Motorway (M) 25), as well as number of data points and basic statistics (Mean, Standard (Std.) Deviation, and Range) of 2010 concentrations (in µg/m³) at each location. Road data source: Meridian

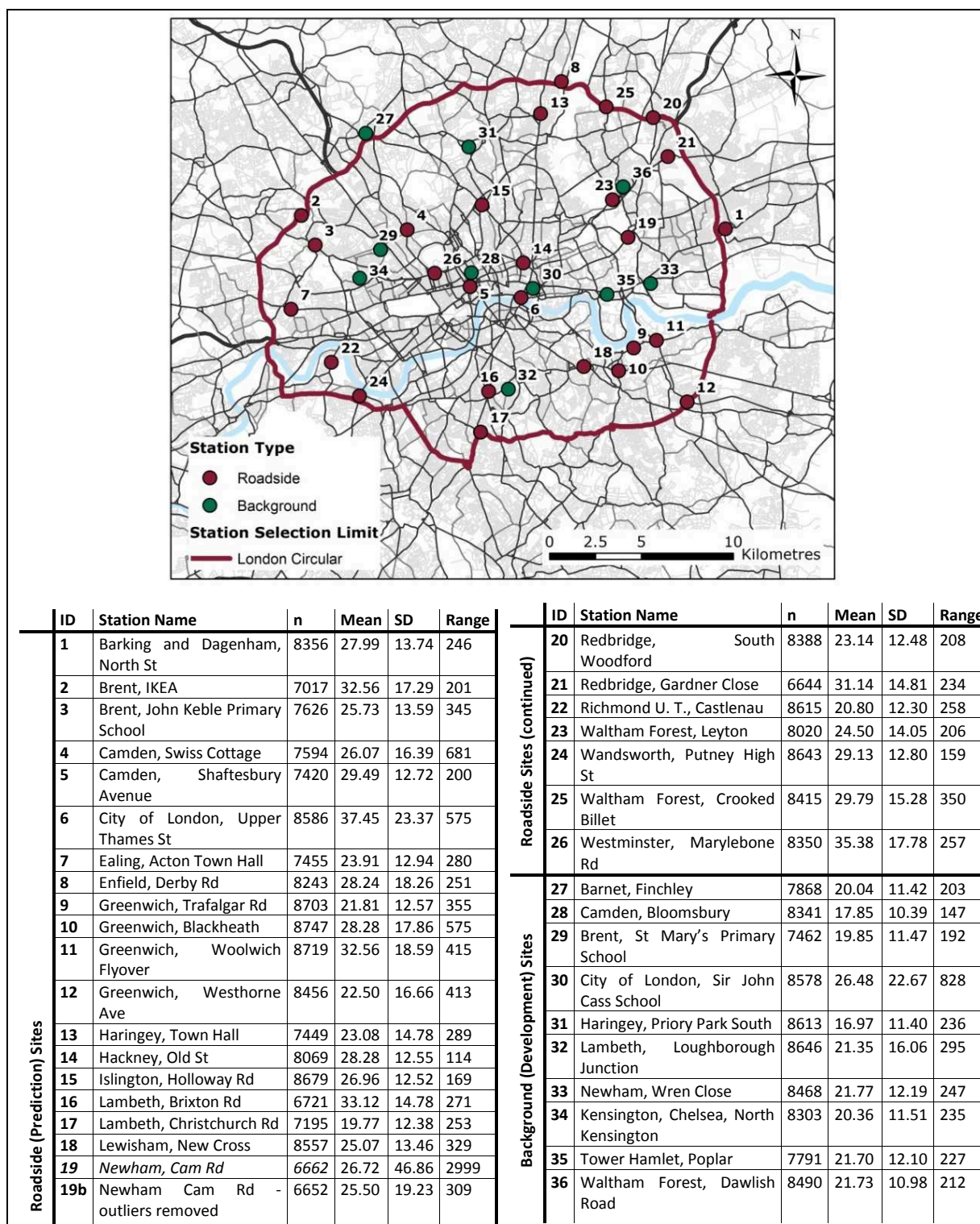


Figure 28: Locations of PM₁₀ monitoring stations in London and limit for station selection (London North and South Circular Road), as well as number of data points and basic statistics (Mean Standard (Std.) Deviation, and Range) of 2010 concentrations (in µg/m³) at each location. Road data source: Meridian

The basic statistics in Figure 27 show that for PM_{2.5} the 2010 mean hourly concentrations range between 11.39µg/m³ and 18.22µg/m³, the standard deviations (SD) range from 7.04µg/m³ to

14.51 $\mu\text{g}/\text{m}^3$ and the range at the different stations lies between 62 $\mu\text{g}/\text{m}^3$ and 439 $\mu\text{g}/\text{m}^3$. Some difference between the station types can be observed between the mean concentrations, which are higher at roadside locations for most of the stations. Differences between roadside and background stations are however not very pronounced.

For PM_{10} the basic statistics presented in Figure 28 show mean hourly concentrations between 19.77 $\mu\text{g}/\text{m}^3$ and 37.45 $\mu\text{g}/\text{m}^3$ for roadside and between 17.85 $\mu\text{g}/\text{m}^3$ and 26.48 $\mu\text{g}/\text{m}^3$ for background stations. The SD ranges between 12.30 $\mu\text{g}/\text{m}^3$ and 23.37 $\mu\text{g}/\text{m}^3$ for roadside and between 10.39 $\mu\text{g}/\text{m}^3$ and 22.67 $\mu\text{g}/\text{m}^3$ for background monitoring stations. The range of mean concentrations and SD are slightly lower for background compared to roadside stations. It can however be observed that some background stations show higher mean and SD compared to some of the roadside stations. The range lies for all stations between 112 $\mu\text{g}/\text{m}^3$ and 828 $\mu\text{g}/\text{m}^3$, no difference could be observed between ranges at roadside and background sites.

Apart from the 2010 concentrations collected at the London monitoring stations described above, data for 2011 was additionally downloaded for London roadside stations to analyse temporal transferability of the temporally adjusted LUR model. Basic statistics (mean, SD, and range) of the 2011 data sets are presented in Appendix H. To test for spatial transferability data was additionally downloaded for network monitoring stations outside London. The selection of the locations was broadly based on the availability of $\text{PM}_{2.5}$ and PM_{10} monitoring stations from areas surrounding London (Thurrock, Stanford-le-Hope; Mole Valley Dorking; Reading New Town; Harwell; Oxford St Ebbes) and for a selection of five further stations of urban locations from different parts of England (Norwich, Bristol, Nottingham, Leeds, Newcastle). Data for all 10 stations was ratified by LAQN or DEFRA and has a data availability above 75% for the relevant time period. Basic statistics (mean, SD, and range) are presented in the Appendix H.

4.4.3. Step 1: Nearest Monitoring Station versus Station Average

As a first step in this analysis, it was tested if variability at a location was better represented by the nearest background station or the average of background station concentrations (using development sites). As mentioned in chapter 2, locations have commonly been predicted by using concentrations from the nearest monitor or from an average between several stations in the area. The question whether one or the other approach works better has however not been answered and is to a large extent also dependent on study area (geographical scale) and degree of difference between the location types. For example any two stations in London are probably better correlated

with another than with a station in Brighton, but a London background station may be less well correlated to a close-by roadside station than to another background station a bit further away.

In order to compare the two approaches, Pearson's correlation was applied between the hourly monitoring concentrations at each station and 1) the nearest background monitoring station or 2) an average of all (other) background stations. In preparation for approach 1, the nearest background site was identified in a GIS. For approach 2, the hourly, daily, and weekly average of all development sites was calculated for correlation with prediction sites. The hourly average of all prediction sites for PM_{2.5} had seven missing data points. All other datasets for approach 2 showed no missing data. In order to also apply approach 2 to development sites, the average of all other development sites was prepared.

	ID	Nearest ID	Hourly		Daily		Weekly	
			r: Nearest	r: Average	r: Nearest	r: Average	r: Nearest	r: Average
Roadside (Prediction) Sites	1	12	0.655	0.659	0.704	0.732	0.718	0.750
	2	12	0.809	0.790	0.859	0.843	0.871	0.824
	3	10	0.802	0.844	0.883	0.942	0.899	0.941
	4	10	0.790	0.860	0.880	0.940	0.911	0.944
	5	10	0.520	0.576	0.552	0.629	0.508	0.552
	6	10	0.848	0.869	0.879	0.923	0.790	0.832
	7	10	0.646	0.686	0.665	0.726	0.435	0.532
	8	10	0.821	0.870	0.920	0.959	0.950	0.974
	9	10	0.869	0.880	0.902	0.942	0.890	0.928
Background (Development) Sites	10	12	0.884	0.896	0.932	0.943	0.916	0.964
	11	12	0.863	0.884	0.939	0.948	0.913	0.961
	12	10	0.884	0.920	0.932	0.972	0.916	0.928
	13	12	0.839	0.857	0.945	0.954	0.905	0.940

Table 35: Pearson's correlation (r) of hourly, daily and weekly PM_{2.5} at network monitoring stations with 1) the nearest background monitoring station (r: Nearest) and 2) the average of all (other) background monitoring stations (r: Average). All correlations are highly significant (at a 0.01 level). Correlation result that are higher for approach 1 or 2 by more than 0.01 are presented in blue and bold.

The correlation results for PM_{2.5} are presented in Table 35 with higher correlation (by at least 0.01) between approach 1 and 2 for the results marked in blue. For hourly concentrations Pearson's correlations (r) for approach 1, using the nearest station, range from 0.520 to 0.884. Results for approach 2, using the average, range from 0.576 to 0.920. Only one out of 13 stations showed higher Pearson's correlation by more than 0.01 for approach 1. For daily averages Pearson's correlation ranges from 0.552 to 0.945 for approach 1 (nearest) and from 0.629 to 0.972 for approach 2 (average). For 11 out of 13 stations approach 2 shows higher correlation results by more than 0.01 compared to approach 1. For weekly concentrations Pearson's correlation for approach 1 ranged from 0.435 to 0.916 and for approach 2 from 0.532 to 0.974. 12 out of 13 stations show a

higher correlation for approach 2 (average). The lowest Pearson's correlation for both approaches and all time steps (hourly, daily, weekly) can be observed at station 5 and station 7. For station 5 the correlations vary between 0.508 and 0.629 between different approaches and temporal aggregations. For station 7 correlations vary between 0.435 and 0.726 for different approaches and temporal aggregations. Some difference could be observed between correlations for background (development) and for roadside (prediction) sites. Correlations between background stations (development sites) show some of the highest correlation results for both approaches and all levels of temporal aggregation. In comparison, several of the prediction sites show lower correlations, which most likely reflects the higher local variability at roadside locations.

Results for PM_{10} are presented in Table 36. Pearson's correlations for hourly concentrations have a wide range from 0.266 to 0.913 for approach 1 (near), and from 0.400 to 0.904 for approach 2 (average). For six stations the correlations are higher (by more than 0.01) when using approach 1, 23 stations show higher correlations (by more than 0.01) for approach 2. For daily concentrations, results for Pearson's correlation range between 0.437 and 0.970 for approach 1 and from 0.584 to 0.967 for approach 2. 24 out of all 36 stations show a higher correlation for approach 2 by more than 0.01. For weekly averages the results for Pearson's correlation are between 0.207 and 0.967 for approach 1, and between 0.488 and 0.962 for approach 2. 21 out of 36 stations showed higher correlation results for approach 2. Most stations showed high correlations ($r > 0.7$) for all levels of temporal aggregation (hourly, daily, and weekly) and both approaches. Some of the highest correlations could be observed at several development sites (e.g. site 34 and 35). A few stations however showed lower correlations. Some of the lowest correlations could be observed for station 30, a development site with correlations ranging from 0.320 to 0.666 (for different approaches and levels of temporal aggregation). Station 30 has been identified as "nearest station" for several of the other stations, some of which in turn show low correlations for approach 1 (nearest). Station 6 has station 30 as its nearest background monitor and shows particularly low correlation for approach 1 of 0.266 for hourly, 0.437 for daily and 0.207 for weekly concentrations. Station 6 in addition however shows comparably low correlation results for approach 2 with 0.496 for hourly, 0.655 for daily and 0.606 for weekly concentrations.

	ID	Near ID	Hourly		Daily		Weekly	
			r: Nearest	r: Average	r: Nearest	r: Average	r: Nearest	r: Average
Roadside (Prediction) Sites	1	33	0.794	0.844	0.901	0.924	0.818	0.853
	2	34	0.718	0.723	0.858	0.846	0.829	0.821
	3	34	0.862	0.853	0.926	0.917	0.906	0.864
	4	29	0.658	0.675	0.846	0.851	0.857	0.875
	5	28	0.609	0.712	0.637	0.815	0.471	0.748
	6	30	0.266	0.496	0.437	0.655	0.207	0.606
	7	34	0.892	0.883	0.949	0.943	0.937	0.921
	8	31	0.576	0.666	0.711	0.779	0.548	0.647
	9	35	0.871	0.873	0.944	0.947	0.933	0.939
	10	35	0.773	0.795	0.917	0.931	0.924	0.930
	11	33	0.658	0.639	0.756	0.705	0.746	0.677
	12	33	0.788	0.839	0.564	0.584	0.551	0.565
	13	31	0.689	0.798	0.804	0.902	0.673	0.885
	14	30	0.361	0.837	0.604	0.895	0.378	0.766
	15	31	0.705	0.849	0.826	0.911	0.738	0.887
	16	32	0.657	0.792	0.767	0.851	0.663	0.760
	17	32	0.644	0.749	0.817	0.846	0.768	0.754
	18	35	0.819	0.821	0.896	0.894	0.897	0.893
	19	36	0.216	0.221	0.472	0.478	0.455	0.469
	19b	36	0.550	0.565	0.736	0.754	0.661	0.671
	20	36	0.809	0.804	0.891	0.898	0.810	0.840
	21	34	0.739	0.788	0.800	0.843	0.612	0.698
	22	36	0.797	0.842	0.921	0.944	0.921	0.937
	23	34	0.781	0.829	0.884	0.904	0.839	0.840
	24	36	0.627	0.659	0.726	0.731	0.662	0.636
	25	28	0.630	0.805	0.727	0.898	0.628	0.883
	26	33	0.569	0.611	0.606	0.629	0.462	0.488
Background (Development)	27	31	0.753	0.871	0.854	0.953	0.743	0.941
	28	30	0.320	0.785	0.549	0.834	0.394	0.688
	29	34	0.913	0.883	0.970	0.947	0.967	0.923
	30	28	0.320	0.400	0.549	0.666	0.394	0.516
	31	27	0.753	0.785	0.854	0.911	0.743	0.847
	32	30	0.288	0.697	0.559	0.846	0.470	0.801
	33	35	0.878	0.859	0.941	0.945	0.919	0.919
	34	29	0.913	0.896	0.970	0.966	0.967	0.962
	35	33	0.878	0.904	0.941	0.960	0.919	0.950
	36	33	0.856	0.890	0.950	0.947	0.954	0.932

Table 36: Pearson correlation of hourly PM₁₀ at network monitoring stations with 1) the nearest background monitoring station (r Nearest) and 2) the average of all (other) background monitoring stations (r Average). Correlation result that are higher by more than 0.01 are presented in blue and bold. ** Significant on a 0.01 level

In summary, results reveal that a few monitoring stations are not very well correlated with other stations. Some of these stations are examined in more detail in the following paragraphs. For PM_{2.5}, two stations in particular showed low correlation results: station 5 (Greenwich, Woolwich Flyover) and station 7 (Hackney, Old St). Pictures of both stations are presented in Figure 29. The picture of station 5 (Figure 29, A) reveals two likely reasons for the comparably low correlations with other stations: 1) The station is located very close to the kerb (about 1.50m away), and 2) the station is located at a major road junction, next to a roundabout, as well as under a road flyover. These local details make it likely that station 5 is influenced by high localised concentration variability due to

proximity and amount of traffic sources around the site. Station 7 (Figure 29, B) is located next to a busy five lane road, apart from this general location (which is similar to other, better correlated stations) no specific features of the immediate environment could be identified, which may have explained in comparison lower correlations with other stations.



Figure 29: Street view of network monitoring stations: A) Greenwich - Woolwich Flyover (station ID 5), and B) Hackney – Old St (Station ID 7) with locations of each station indicated. Source: Google maps (<https://www.google.com/maps> accessed: 23rd April 2014).

For PM_{10} two stations have also showed comparably low correlations with the other (background) stations: Station 6 (City of London – Upper Thames St) and station 30 (City of London – Sir John Cass School). Both stations are located in central London (see Figure 28). The site picture for station 6 reveals a very particular location under a bridge (see Figure 30, A). This creates confined conditions with potential accumulation of particles as for example observed for tunnels (Svartengren et al. 2000; Giugliano et al. 2005) and may have created conditions different from locations of other monitoring stations in London.

The second station, which showed comparably low correlation with other stations for PM_{10} , is station 30 (Figure 30, B), a background station. The picture of the station shows that it is located in a quiet courtyard. The location is however very central and as such relatively close to major roads (closest major road less than 50m distance). The proximity to major roads may have had an influence on the particle concentrations at this location. In addition, the high continuous walls around the courtyard may have created specific wind pattern, which could explain why this monitoring station shows different concentration variability compared to other stations with similar proximity to a major roads.

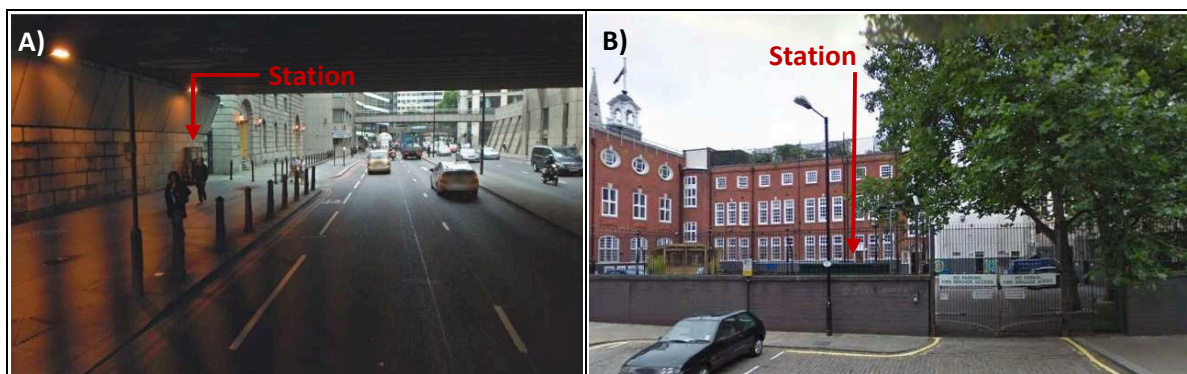


Figure 30: Street view of network monitoring stations: A) City of London - Thames Link Road (Station ID 6), and B) City of London – Sir John Cass School (station ID 30) with locations of each monitoring station indicated. Source: Google maps (<https://maps.google.co.uk>; accessed: 7th Feb. 2014).

Differences in correlation results between approach 1 and 2 show that most stations have a higher correlation with the station average (approach 2) compared to the nearest background station, both for $PM_{2.5}$ and PM_{10} and for all levels of temporal aggregation. For further applications (steps 2, 3, and 4) a decision was therefore made to calculate adjustments for the LUR model using approach 2, the averages of all background stations in the area.

4.4.4. Step 2: Building Temporal Adjustments for LUR Model Predictions

In this section concentrations from development sites were used to develop two different versions of hourly adjustments for LUR predictions of annual mean concentrations for 2010. Applied adjustments include A) direct adjustment by monitoring data, and B) adjustments calculated from monitoring data aggregated by temporal pattern. As mentioned above both of these types of adjustments have been used in the literature. Little comparison between the methods or evaluation with ambient monitoring data had however been undertaken. Adjustment version A has been calculated to evaluate to what extent hourly variability at background stations reflects variability at other locations, and whether it therefore can be used for predictions. Adjustment version B uses temporal trends that reflect certain temporal changes in PM concentrations. This is considered a more transferable method as the repeated pattern can be used to predict concentrations for different time periods or areas for which monitoring data might not be available. In the following paragraphs describe how the two adjustments are calculated.

Development of temporal adjustments:

For adjustment A, the average of all background stations (as described above) was divided directly by the annual mean of $14.08\mu\text{g}/\text{m}^3$ for $\text{PM}_{2.5}$ and $20.82\mu\text{g}/\text{m}^3$ for PM_{10} . The resulting ratios are presented in Figure 31. The two graphs for hourly ratios at the top of Figure 31 show that calculating ratios directly from the hourly background concentrations results in an adjustment ratio with high variability from 0.15 up to 10.33 for $\text{PM}_{2.5}$ and from 0.11 to 8.56 for PM_{10} .

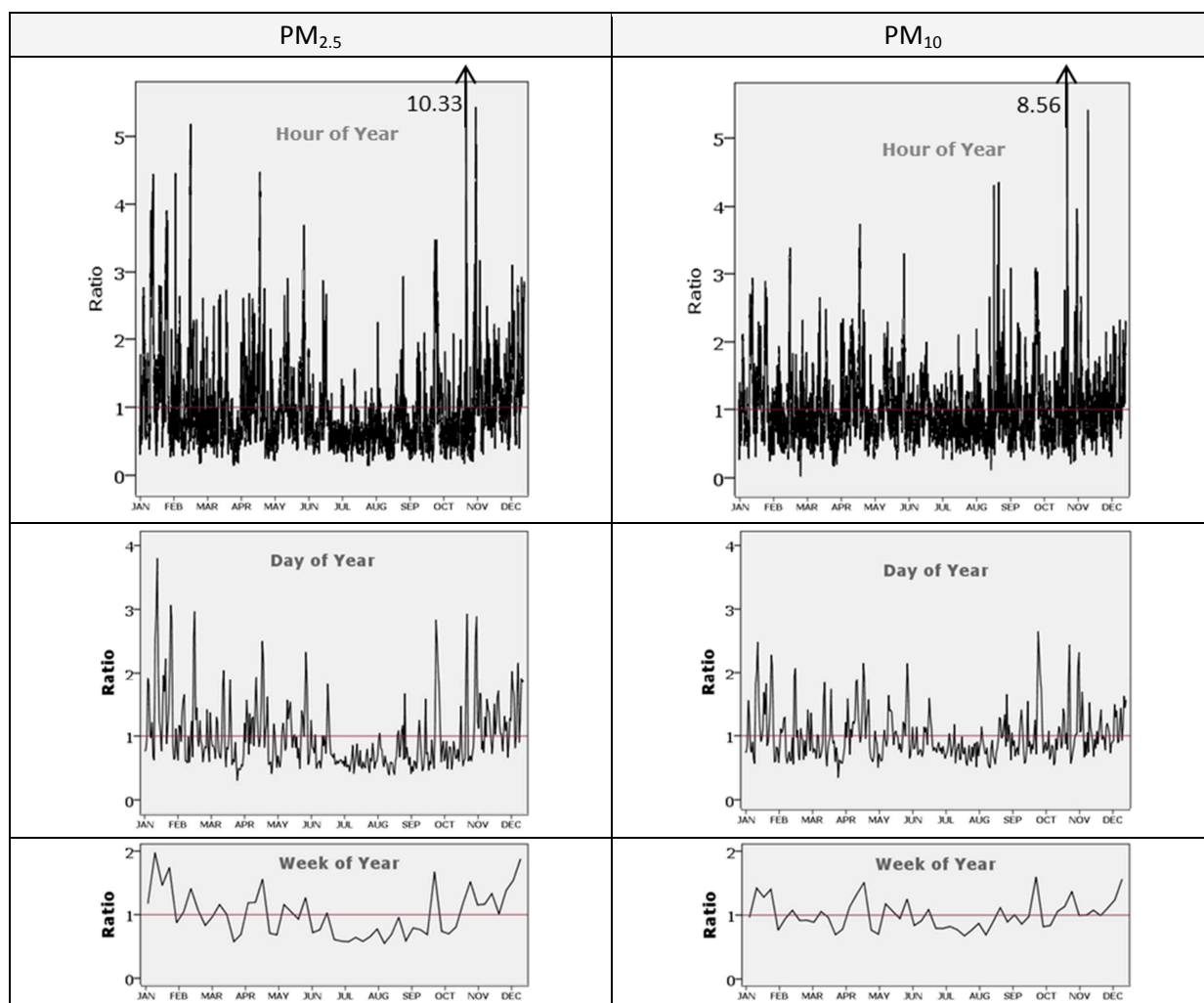


Figure 31: Temporal adjustments A for hourly, daily and weekly LUR predictions of $\text{PM}_{2.5}$ and PM_{10} , based on 2010 data.

The highest ratios both for $\text{PM}_{2.5}$ and PM_{10} have been calculated for a short time period in November. These values reflect a pollution episode referred to by the LAQN as the 'Guy Fawkes and Diwali event celebrations' (www.londonair.org.uk). Ratios for all other times remain below 6 for both $\text{PM}_{2.5}$ and PM_{10} with the majority of hourly ratios between 0 and 2. For daily ratios (graphs in the middle of Figure 31), peaks are less pronounced compared to hourly ratios. All ratios are

between 0 and 4 for $PM_{2.5}$ and between 0 and 3 for PM_{10} . For weekly ratios (Figure 31, bottom) a further reduction in variability can be observed with all ratios between 0 and 2 both for $PM_{2.5}$ and for PM_{10} .

Next, adjustment B was calculated. As a first step, temporal patterns were identified, which describe repeated patterns of pollution variability. Hour of day and day of the week form recognised patterns in particle pollution following differences in traffic source activities (Beevers et al. 2009). The third recognised temporal pattern for particle variability is season. Seasonal differences have been described as part of monthly differences, as well as differences between winter, spring, summer and autumn (Arx et al. 2004; Beevers et al. 2009). Seasonal differences in particle concentrations reflect climatological changes throughout the year.

For this application hour of day, day of week and seasonal patterns were combined to calculate adjustment B. For the seasonal pattern four seasons were used instead of the monthly difference, as weather, and as a consequence related particle concentration changes, can vary substantially between the same months of different years. Differences between the four seasons are more stable between years. As a first step the hourly monitoring concentrations for the average of London background stations were used to calculate the mean concentrations by hour of day, day of week and season. These three averages were then each divided by the 2010 annual mean. The resulting ratios were combined to adjustments B, as presented in Figure 32. To calculate hourly ratios, first the seasonal ratio was multiplied by the day of the week, then the hour of day was combined with the results. All resulting ratios of adjustment B are presented in Appendix H. In comparison to the direct hourly adjustment A (Figure 31) the graphs show a more repetitive pattern, as well as much less variability with the highest ratios well below 2 for both $PM_{2.5}$ and PM_{10} . For the daily adjustment B, only the seasonal and day of the week ratios were combined. Weeks were adjusted only by seasonal ratios. Ratios for adjustment versions A and B were subsequently multiplied with ESCAPE-LUR estimates, calculated for each station of the prediction monitoring sites.

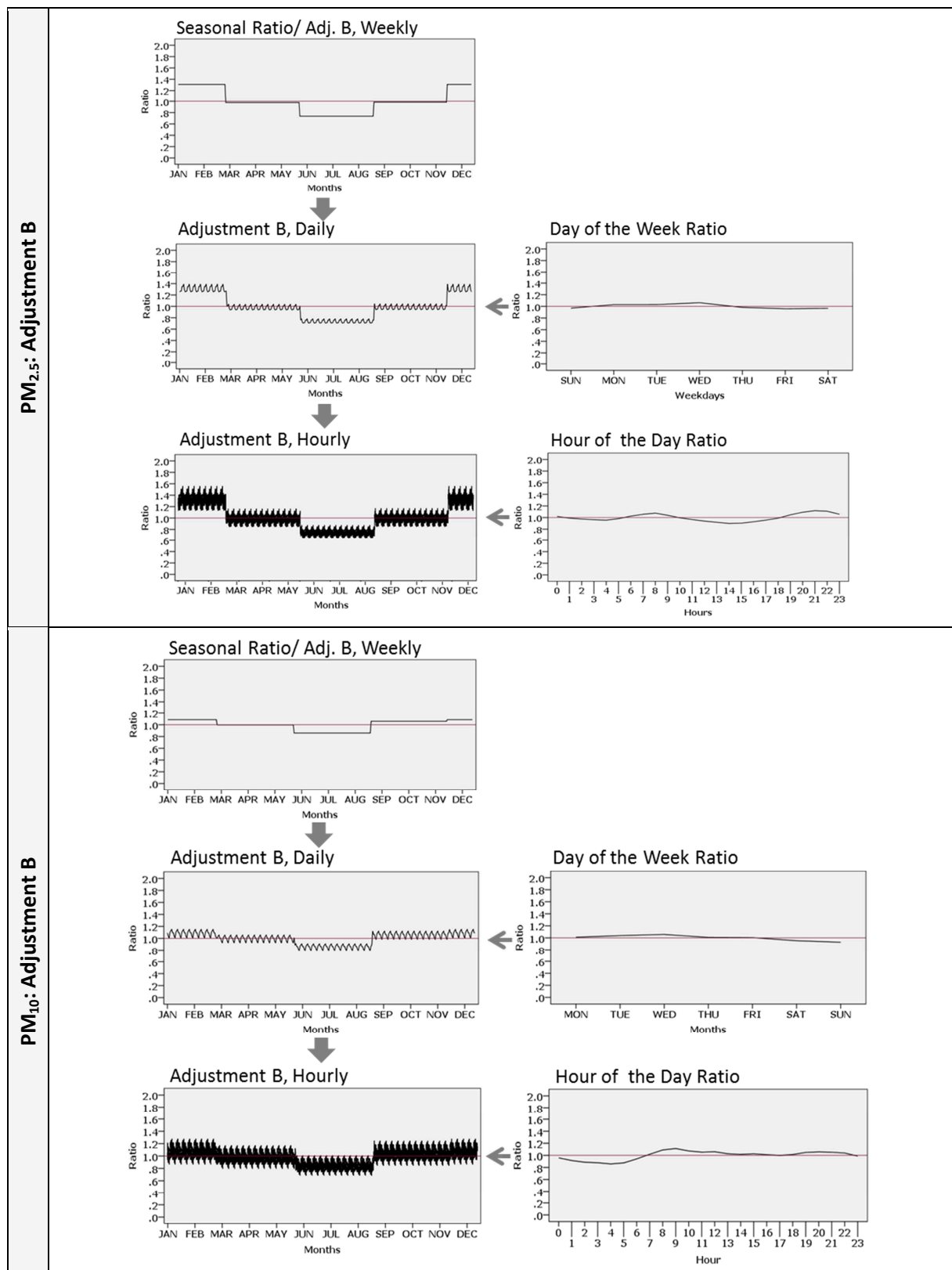


Figure 32: Temporal adjustments B of hourly, daily and weekly LUR predictions of PM_{2.5} and PM₁₀, based on 2010 data.

Adjustment by ratio and adjustment by total difference in comparison

Applying ratios (and in particular combining several ratios) to a dataset can potentially provide a scaling problem as the same ratio adds more to a high value compared to a lower value. A different approach is to calculate and apply the direct difference to a long term mean. Adjusting by total difference to a long term mean would add the same value to a low and a high concentration. Adjustment by difference can however result in negative values, ratio adjustment is therefore preferred here, as well as by many other studies (Aguilera et al. 2009; Aguilera et al. 2010; Brauer et al. 2008; Dons, Van Poppel, Int Panis, et al. 2014) as a method. In order to test if the adjustment produces problems with scaling, an adjustment by difference has been calculated both for adjustment A and B and is compared in this section.

In order to calculate adjustment A using difference, the average (hourly, daily, and weekly) for all background monitoring stations is taken minus the annual average. Similarly, adjustment B is developed using total difference to the annual average for hour of day, day of week, and season. These three differences are then added together (as shown in Figure 32). To calculate adjusted LUR estimates the adjustments by difference are added to LUR estimates of the prediction sites. Predictions of monitoring stations were compared between both adjustment variations to assess if adjustment by ratio performed less well, which would indicate problems with its application.

For comparison between ratio and difference adjustments COD (Coefficient of Divergence) was calculated between monitored and modelled. COD has been introduced in chapter 3. A COD of 0 means values are the same, while a Pearson correlation of 1 refers to a perfect correlation (Gaines Wilson & Zawar-reza 2006). The COD is more suited than Pearson's correlation to identify a potential problem of scaling as it compares not only the variability, but also the total values.

COD results for $PM_{2.5}$ are presented in Table 37. Result show that most COD differ little between adjustment by ratio and by total difference for both adjustments A and B, as well as for different levels of temporal aggregation. Only for weekly concentrations at one station (station 5) for adjustment B the difference in COD between the two approaches was higher than 0.05. Furthermore, there is no clear indication if adjustment by ratio or by difference generally show better COD results. For both adjustment A and B, as well as for all periods of temporal aggregation, COD are lower at a relatively equal number of stations for either adjustment. In summary, the results for $PM_{2.5}$ show no clear evidence that one or the other version of adjustment has better agreement with concentrations at the different stations.

Station ID	Hourly				Daily				Weekly			
	Adjustment A		Adjustment B		Adjustment A		Adjustment B		Adjustment A		Adjustment B	
	Ratio	Diff.	Ratio	Diff.	Ratio	Diff.	Ratio	Diff.	Ratio	Diff.	Ratio	Diff.
1	0.216	0.205	0.265	0.255	0.160	0.147	0.199	0.188	0.126	0.111	0.119	0.104
2	0.181	0.190	0.280	0.254	0.134	0.142	0.229	0.202	0.105	0.112	0.174	0.135
3	0.173	0.187	0.292	0.279	0.111	0.123	0.238	0.226	0.101	0.106	0.184	0.169
4	0.167	0.166	0.293	0.282	0.112	0.112	0.241	0.229	0.104	0.104	0.176	0.161
5	0.287	0.320	0.240	0.271	0.251	0.279	0.182	0.226	0.215	0.236	0.134	0.188
6	0.302	0.337	0.254	0.299	0.279	0.303	0.237	0.285	0.269	0.282	0.229	0.277
7	0.223	0.209	0.251	0.245	0.180	0.167	0.199	0.197	0.155	0.143	0.138	0.136
8	0.219	0.259	0.257	0.266	0.200	0.227	0.222	0.239	0.193	0.207	0.197	0.215
9	0.173	0.186	0.295	0.281	0.108	0.123	0.240	0.223	0.080	0.079	0.165	0.137

Table 37: COD (Coefficient of Divergence) for adjustments by ratio and difference of the temporal LUR adjustment versions A (direct adjustment by monitoring data) and B (adjustment based on temporal patterns) for hourly, daily, and weekly PM_{2.5} concentrations. Lower COD results between ratio and difference by more than 0.01 are coloured in blue and bold.

ID	Hourly				Daily				Weekly			
	Adjustment A		Adjustment B		Adjustment A		Adjustment B		Adjustment A		Adjustment B	
	Ratio	Diff.	Ratio	Diff.	Ratio	Diff.	Ratio	Diff.	Ratio	Diff.	Ratio	Diff.
1	0.267	0.299	0.258	0.281	0.250	0.268	0.234	0.261	0.248	0.255	0.227	0.254
2	0.169	0.164	0.235	0.226	0.117	0.113	0.178	0.175	0.098	0.095	0.123	0.122
3	0.121	0.128	0.231	0.220	0.082	0.085	0.178	0.168	0.065	0.064	0.117	0.104
4	0.193	0.211	0.290	0.274	0.128	0.144	0.220	0.205	0.097	0.108	0.158	0.140
5	0.200	0.192	0.209	0.212	0.175	0.169	0.165	0.181	0.160	0.158	0.139	0.160
6	0.223	0.207	0.241	0.237	0.200	0.185	0.192	0.198	0.174	0.164	0.151	0.164
7	0.105	0.108	0.228	0.212	0.062	0.068	0.179	0.164	0.046	0.053	0.127	0.108
8	0.221	0.221	0.266	0.267	0.181	0.181	0.202	0.214	0.170	0.170	0.161	0.181
9	0.107	0.108	0.235	0.219	0.068	0.070	0.187	0.173	0.051	0.055	0.132	0.115
10	0.153	0.147	0.226	0.221	0.120	0.116	0.177	0.176	0.112	0.110	0.132	0.135
11	0.196	0.183	0.228	0.224	0.167	0.153	0.171	0.175	0.138	0.129	0.117	0.130
12	0.168	0.179	0.296	0.282	0.119	0.130	0.241	0.226	0.115	0.120	0.180	0.161
13	0.144	0.144	0.254	0.241	0.098	0.098	0.195	0.183	0.067	0.067	0.132	0.117
14	0.148	0.135	0.204	0.200	0.115	0.104	0.152	0.156	0.101	0.095	0.107	0.117
15	0.120	0.125	0.222	0.208	0.081	0.081	0.164	0.153	0.061	0.057	0.104	0.089
16	0.164	0.140	0.198	0.193	0.119	0.096	0.144	0.142	0.085	0.068	0.086	0.085
17	0.219	0.218	0.287	0.269	0.129	0.129	0.205	0.183	0.092	0.092	0.146	0.114
18	0.140	0.150	0.244	0.230	0.100	0.108	0.192	0.179	0.078	0.084	0.137	0.120
19	0.184	0.185	0.269	0.256	0.151	0.148	0.195	0.190	0.134	0.131	0.140	0.137
19b	0.181	0.182	0.267	0.253	0.132	0.128	0.182	0.175	0.105	0.102	0.120	0.114
20	0.228	0.342	0.244	0.258	0.200	0.230	0.202	0.223	0.192	0.204	0.177	0.202
21	0.154	0.136	0.203	0.203	0.110	0.099	0.157	0.158	0.088	0.079	0.098	0.100
22	0.135	0.145	0.265	0.248	0.098	0.106	0.213	0.197	0.086	0.089	0.155	0.135
23	0.207	0.242	0.242	0.250	0.191	0.209	0.204	0.219	0.186	0.194	0.171	0.194
24	0.173	0.156	0.208	0.199	0.119	0.097	0.138	0.135	0.095	0.085	0.107	0.104
25	0.296	0.331	0.281	0.294	0.287	0.306	0.261	0.284	0.280	0.287	0.257	0.280
26	0.198	0.190	0.240	0.234	0.164	0.152	0.182	0.182	0.120	0.103	0.102	0.108

Table 38: COD (Coefficient of Divergence) for adjustments by ratio and difference of the temporal LUR adjustment A (direct adjustment by monitoring data) and B (adjustment based on temporal patterns) for hourly, daily, and weekly PM₁₀ concentrations. Lower COD results between ratio and difference by more than 0.01 are coloured in blue and bold.

Results for PM_{10} are presented in Table 38. Results show that at a large number of stations COD differs by less than 0.01 between adjustments by ratio and by total difference. For adjustment A both for hourly, as well as for daily concentrations, COD differ by less than 0.01 at 14 out of 26 stations. Of all comparisons between ratio and difference adjustments presented in the table only 19 COD results differ by more than 0.02, and only for one comparison COD results differ by more than 0.05 (hourly, adjustment A, station 20). In addition, results do not show consistently better agreement (lower COD) with concentrations at monitoring stations for one or the other adjustment version. In summary, for PM_{10} (as above for $PM_{2.5}$) the comparison between predictions by ratio and difference both for adjustments A and B, as well as for all levels of temporal aggregation show very similar results.

For both PM size fractions, the adjustment by ratio did not, as such show any lower performance in comparison with application of the total difference. As mentioned before, adjustment by difference can produce negative concentrations. Ratio adjustments were therefore chosen over adjustment by total difference and have been used in steps three and four of this analysis.

4.4.5. Step 3: Comparing Temporal Adjustments

In the following section the two versions of temporal adjustment A (direct adjustment) and B (adjustment by temporal patterns), as developed above, are compared in detail. The comparison is made by assessing how well each approach predicts variability of the set of prediction sites, using Pearson's correlation. First, predictions for 2010 of London roadside stations are assessed, then concentrations for year 2011 are predicted for the same stations (dependent on data availability for year 2011) to test to what extent the adjustments would be temporally transferable. Subsequently predictions are made for concentrations at 10 monitoring stations outside London to test for spatial transferability of the approaches.

Predicting monitoring stations in London for 2010

Temporal adjustments A and B have been applied to LUR estimates of the set of prediction sites, as described above. The ability of the two adjustment versions to reflect concentration variability was examined and compared using Pearson's correlation. Results are presented in Table 39 for $PM_{2.5}$ and in Table 40 for PM_{10} . For both $PM_{2.5}$ and PM_{10} and for all levels of temporal aggregation the correlations were lower for adjustment B compared to adjustment A at every station. For hourly concentrations of $PM_{2.5}$, correlations for adjustment A range from 0.576 to 0.880 compared to

between 0.063 and 0.274 for adjustment B. For hourly PM₁₀ concentrations a similar difference was observed. Correlations range from 0.496 to 0.883 for adjustment A and from 0.130 to 0.317 for adjustment B. A similar difference was also observed between daily adjustments with the results of correlations for adjustment A higher than for adjustment B, for PM_{2.5} (adj. A: 0.629 < r > 0.942, adj. B: 0.170 < r > 0.355) and for PM₁₀ (adj. A: 0.584 < r > 0.947, adj. B: 0.094 < r > 0.368). For weekly concentrations the same difference could as well be observed between adjustments A and B for PM_{2.5} (adj. A: 0.552 < r > 0.974, adj. B: 0.097 < r > 0.587), as well as for PM₁₀ (adj. A: 0.488 < r > 0.939, adj. B: 0.048 < r > 0.625). The conclusion was drawn that temporally adjusted LUR predictions using adjustment A better reflected variability at other locations compared to adjustment B when applied to concentrations of the same time period and area.

ID	2010 Data						2011 Data					
	Hourly		Daily		Weekly		Hourly		Daily		Weekly	
	Adj. A	Adj. B	Adj. A	Adj. B	Adj. A	Adj. B	Adj. A	Adj. B	Adj. A	Adj. B	Adj. A	Adj. B
1	0.659**	0.138**	0.732**	0.206**	0.750**	0.397**	0.037**	0.093**	0.098	0.106	0.006	0.098
2	0.790**	0.231**	0.843**	0.320**	0.824**	0.541**	-	-	-	-	-	-
3	0.844**	0.252**	0.942**	0.336**	0.941**	0.551**	0.082**	0.108**	0.146**	0.107*	0.101	0.162
4	0.860**	0.220**	0.940**	0.307**	0.944**	0.542**	-	-	-	-	-	-
5	0.576**	0.093**	0.629**	0.170**	0.552**	0.368**	-0.011	0.049**	0.037	0.061	-0.131	0.027
6	0.869**	0.134**	0.923**	0.190**	0.832**	0.326**	-	-	-	-	-	-
7	0.686**	0.063**	0.726**	0.039**	0.532**	0.097	0.020	0.048**	0.074	0.014	-0.056	-0.007
8	0.870**	0.274**	0.959**	0.355**	0.974**	0.587**	-	-	-	-	-	-
9	0.880**	0.222**	0.942**	0.302**	0.928**	0.531**	0.048**	0.064**	0.104*	0.064	0.042	0.065

Table 39: Pearson's correlations (r) between monitored concentrations and the temporally adjusted LUR model using adjustment A (direct adjustment by monitoring data) and B (adjustment based on temporal patterns) for hourly, daily and weekly PM_{2.5} predictions of London roadside stations for 2010 and 2011. * Significant on a 0.05 level, ** Significant on a 0.01 level

Testing temporal transferability

The construction of adjustment B was aimed to reflect transferable patterns in pollution which can be applied directly to predict different years or locations. Temporal transferability is tested here for 2011 concentrations for the same prediction sites. Data was however not available for all stations and only five out of nine PM_{2.5} stations and 20 out of 26 PM₁₀ stations had sufficient data availability. The temporally adjusted LUR predictions based on 2010 data (adjustments A and B) were directly compared to 2011 concentrations. Adjustment A is added to this comparison mostly as a reference to show if in comparison adjustment B reflects a transferable temporal patterns. Direct adjustment with monitoring data as used for adjustment A would usually be calculated from 2011 background concentrations to predict locations for 2011.

Results for Pearson's correlations between monitored and modelled are presented in Table 39 (right side) for PM_{2.5} and in Table 40 (right side) for PM₁₀. For PM_{2.5} all correlation results for 2011 are below 0.2 and only station 3 shows highly significant results (at a 0.01 level) above a correlation of 0.1 for hourly concentrations for adjustment B, and for daily concentrations both for adjustments A and B.

ID	2010 Data						2011 Data					
	Hourly		Daily		Weekly		Hourly		Daily		Weekly	
	Adj. A	Adj. B	Adj. A	Adj. B	Adj. A	Adj. B	Adj. A	Adj. B	Adj. A	Adj. B	Adj. A	Adj. B
1	0.844**	0.139**	0.924**	0.134**	0.853**	0.214	-	-	-	-	-	-
2	0.723**	0.305**	0.846**	0.313**	0.821**	0.461**	0.072**	0.266**	0.105	0.264**	0.046	0.298*
3	0.853**	0.169**	0.917**	0.183**	0.864**	0.254	0.082**	0.220**	0.117*	0.234**	0.102	0.280
4	0.675**	0.288**	0.851**	0.325**	0.875**	0.503**	0.063**	0.242**	0.085	0.248**	0.039	0.293*
5	0.712**	0.239**	0.815**	0.094	0.748**	0.108	0.075**	0.175**	0.092	0.072	-0.037	0.010
6	0.496**	0.295**	0.655**	0.297**	0.606**	0.347**	0.053**	0.274**	0.052	0.211**	-0.068	0.185
7	0.883**	0.253**	0.943**	0.331**	0.921**	0.503**	0.039**	0.164**	0.074	0.145**	-0.020	0.170
8	0.666**	0.185**	0.779**	0.124**	0.647**	0.048	0.045**	0.165**	0.070	0.146**	0.124	0.194
9	0.873**	0.218**	0.947**	0.256**	0.939**	0.425**	0.077**	0.156**	0.127*	0.154**	0.073	0.186
10	0.795**	0.226**	0.931**	0.312**	0.930**	0.521**	0.060**	0.176**	0.083	0.155**	0.036	0.171
11	0.639**	0.247**	0.705**	0.243**	0.677**	0.337**	0.030**	0.234**	0.029	0.209**	-0.065	0.181
12	0.839**	0.183**	0.584**	0.180**	0.565**	0.412**	0.042**	0.123**	0.077	0.145**	-0.003	0.172
13	0.798**	0.220**	0.902**	0.295**	0.885**	0.464**	-	-	-	-	-	-
14	0.837**	0.229**	0.895**	0.149**	0.766**	0.147	0.114**	0.267**	0.165**	0.249**	0.073	0.241
15	0.849**	0.268**	0.911**	0.242**	0.887**	0.361**	0.033**	0.257**	0.047	0.239**	-0.025	0.262
16	0.792**	0.185**	0.851**	0.200**	0.760**	0.335*	0.052**	0.097**	0.072	0.034	-0.026	-0.019
17	0.749**	0.263**	0.846**	0.368**	0.754**	0.625**	-	-	-	-	-	-
18	0.821**	0.237**	0.894**	0.259**	0.893**	0.442**	0.080**	0.194**	.131*	0.165**	0.104	0.198
19	0.221**	0.107**	0.478**	0.173**	0.469**	0.344*	-	-	-	-	-	-
19b	0.565**	0.248**	0.754**	0.288**	0.671**	0.444**	-	-	-	-	-	-
20	0.804**	0.179**	0.898**	0.206**	0.840**	0.320*	0.048**	0.220**	.104*	0.316**	0.091	0.397**
21	0.788**	0.130**	0.843**	0.104**	0.698**	0.242	0.107**	0.402**	.186**	0.512**	0.212	0.665**
22	0.842**	0.214**	0.944**	0.246**	0.937**	0.415**	0.050**	0.164**	0.090	0.153**	0.027	0.185
23	0.829**	0.188**	0.904**	0.218**	0.840**	0.300*	-	-	-	-	-	-
24	0.659**	0.247**	0.731**	0.169**	0.636**	0.270	0.078**	0.241**	.111*	0.165**	0.059	0.167
25	0.805**	0.317**	0.898**	0.291**	0.883**	0.481**	-	-	-	-	-	-
26	0.611**	0.253**	0.629**	0.174**	0.488**	0.171	0.037**	0.335**	0.021	0.273**	-0.056	0.305*

Table 40: Pearson's correlations (r) between monitored concentrations and the temporally adjusted LUR model using adjustment A (direct adjustment by monitoring data) and B (adjustment based on temporal patterns) for PM₁₀ predictions of London roadside stations for 2010 and 2011. * Significant on a 0.05 level, ** Significant on a 0.01 level

For hourly PM₁₀ concentrations all correlation results are significant at a 0.01 level and adjustment B shows higher correlations compared to adjustment A between 0.097 and 0.402. For daily PM₁₀ concentrations most results for adjustment A are not significant (13 out of 20). Apart from results for two stations, all correlation coefficients for adjustment B are however highly significant (at a 0.01 level) and range from 0.145 to 0.512. Both for hourly, as well as for daily PM₁₀ concentrations correlations with adjustment B are similar and for some stations higher compared to correlations with concentrations for 2010. For weekly PM₁₀ concentrations only correlations at two stations are

highly significant for adjustment B (stations 20 and 21) (No highly significant results for adjustment A). Both stations show higher correlations compared to correlations with data from 2010.

In summary, the application of adjustment B to 2011 concentrations shows for $PM_{2.5}$ little temporal transferability. Results for correlations are very low ($r < 0.2$) and are below correlation results with 2010 data. For PM_{10} adjustment B shows similar and sometimes higher correlation results for 2011 compared to results for 2010 data, indicating good temporal transferability. Temporal adjustment B however only reflects a small part of variability at other stations either for 2010 or 2011 concentrations.

Testing spatial transferability

Both adjustment versions A and B were applied to predict hourly $PM_{2.5}$ and PM_{10} concentrations for year 2010 at 10 locations outside London. The 10 monitoring stations and approximate distance to London are presented in Table 41 showing results for Pearson's correlations between temporally adjusted LUR predictions and monitored concentrations for $PM_{2.5}$ and PM_{10} for year 2010.

All correlations are significant at a 0.01 level. Results show that for both particle size fractions and all levels of temporal aggregation, adjustment A results in higher Pearson's correlations compared to adjustment B. For hourly $PM_{2.5}$ concentrations, correlations range from 0.466 to 0.783 for adjustment A and from 0.206 to 0.363 for adjustment B. For hourly PM_{10} concentrations, correlations range from 0.389 to 0.692 for adjustment A and from 0.156 to 0.236 for adjustment B. The range for daily correlations is a little higher for both adjustments: For daily $PM_{2.5}$ concentrations, results range from 0.593 to 0.926 for adjustment A and from 0.254 to 0.515 for adjustments B. For daily PM_{10} concentrations, correlations range from 0.550 to 0.907 for adjustment A and from 0.227 to 0.348 for adjustment B. Weekly correlations show higher results compared to correlations at the hourly or daily level: For $PM_{2.5}$, correlations range between 0.759 and 0.931 for adjustment A and between 0.454 and 0.711 for adjustment B. For weekly PM_{10} concentrations, correlations range between 0.646 and 0.887 for adjustment A and between 0.358 and 0.497 for adjustment B.

In summary, results for $PM_{2.5}$ show for adjustment A (direct adjustment with monitoring concentrations) a similar range of results as previously presented for stations in London, with only a few stations, such as 'Newcastle Centre' or 'Bristol St Paul's', showing in comparison lower correlations for hourly and daily concentrations. For adjustment B correlations for $PM_{2.5}$ concentrations are generally at least as high and in some cases higher compared to correlations for stations in London. For PM_{10} concentrations, correlation results of adjustment A are for several stations lower than results for stations in London, especially for hourly and daily correlations. For

adjustments B correlations between PM₁₀ concentrations with stations outside London are within the range of results for correlation results of stations inside London for all levels of temporal aggregation.

Station	Dist.	PM _{2.5}						PM ₁₀					
		Hourly		Daily		Weekly		Hourly		Daily		Weekly	
		Adj. A	Adj. B	Adj. A	Adj. B	Adj. A	Adj. B	Adj. A	Adj. B	Adj. A	Adj. B	Adj. A	Adj. B
Thurrock Stanford-le-H.	30	-	-	-	-	-	-	0.784	0.199	0.907	0.276	0.887	0.459
Mole Valley Dorking	35	-	-	-	-	-	-	0.781	0.202	0.907	0.231	0.886	0.422
Reading New Town	55	0.835	0.433	0.926	0.515	0.928	0.711	-	-	-	-	-	-
Harwell	75	0.720	0.293	0.845	0.348	0.901	0.525	0.598	0.154	0.769	0.255	0.753	0.392
Oxford St Ebbes	80	0.783	0.363	0.886	0.426	0.931	0.639	0.692	0.236	0.820	0.348	0.877	0.475
Norwich Lakenfields	150	0.655	0.312	0.779	0.387	0.854	0.613	0.566	0.168	0.739	0.272	0.781	0.382
Bristol St Paul's	170	0.479	0.278	0.634	0.356	0.769	0.605	0.389	0.156	0.550	0.237	0.646	0.453
Nottingham Centre	200	0.651	0.212	0.747	0.260	0.828	0.490	0.498	0.206	0.708	0.321	0.779	0.497
Leeds Centre	300	0.585	0.272	0.699	0.342	0.792	0.565	0.508	0.185	0.643	0.227	0.654	0.358
Newcastle Centre	450	0.466	0.206	0.593	0.254	0.759	0.454	0.436	0.207	0.560	0.260	0.697	0.395

Table 41: Pearson's correlations of temporally adjusted LUR predictions for adjustments A (direct adjustment by monitoring data) and B (adjustment based on temporal patterns) at 10 stations outside London for hourly, daily, and weekly PM_{2.5} and PM₁₀ with approximate distance to Central London given in km. Dist. = Approximate distance from central London. All results are significant on a 0.01 level.

The results show very good spatial transferability for both adjustment approaches with correlations generally similar for locations outside London compared to locations in London. No clear indication of lower correlation coefficients with increasing distance from London could be observed. As for correlations with 2010 concentrations for locations in London, adjustment A was able to explain a much larger part of the concentration variability compared to adjustment B.

Summary

Temporally adjusted LUR concentrations using adjustment version A reflect variability of 2010 concentrations at different locations much better, compared to adjustment B. The variability included in adjustment B is limited to repeated temporal patterns, which only reflect a small part of actual variability. Adjustment B shows generally good spatial transferability and for PM₁₀ as well good temporal transferability. Yet Pearson's correlations remain below 0.5 for all predictions. Adjustment A has shown limited direct temporal transferability. The adjustment can however be

calculated for every year using concentrations from London background monitoring stations (available online, for years since mid-1990s). Using adjustment A therefore provides potential for applications for many years, despite low direct temporal transferability. Spatial transferability was tested and provided good correlation results between modelled and monitored for locations outside London. Considering above results, adjustment version A was selected for further analyses. The LUR model using adjustment version A is referred to as LUR-adj in the following sections.

4.4.6. Step 4: Evaluating Model Performance of LUR-adj Predictions

The above analysis did not yet fully answer the question how well the LUR-adj model performs when predicting ambient PM and how well it compares with other models. For this section the results for the temporally adjusted LUR model (as described above) were therefore compared in more detail to the concentrations at London monitoring stations using model performance parameters. Model performance is evaluated using several parameters: R^2 , Index of Agreement (IA), normalised mean square error (NMSE), and Fractional Bias (FB). These parameters are commonly used to measure model performance and have been introduced in chapter 3. Over- and underprediction as reflected in the FB originate for this analysis in the initial (annual) LUR estimate. The temporal adjustment only modifies the annual average for the different time periods (e.g. days). For PM_{10} , model performance results are in addition calculated for predictions of the ADMS-urban model. Results are then compared to the LUR-adj estimates of each station.

Generally model performance is evaluated for the prediction sited, which are roadside stations. To evaluate model performance as well for background locations the development sites (urban background monitoring stations) were predicted using the LOOCV method. With this method each background station is predicted in turn by applying temporal adjustment calculated with the average of other remaining background stations. This validation tests the stability of the model, as well as evaluates performance for background network monitoring stations.

LUR-adj predictions at roadside stations for $PM_{2.5}$

Model performance of $PM_{2.5}$ predictions is evaluated for the prediction sites, as well as for the development sites. Table 42 shows model performance parameters for hourly, daily, and weekly predictions of 2010 concentration for $PM_{2.5}$. For hourly concentrations R^2 for all different monitoring stations are medium low to medium high ranging from 0.332 to 0.846. The IA shows good model performance for all stations with indices between 0.715 and 0.926. The NMSE ranges between 0.126

and 0.672, and the FB shows both under- and overprediction, ranging between -0.474 and 0.132. For daily results, R^2 range from medium low to high, between 0.396 and 0.946. Daily results for IA range between 0.720 and 0.950, showing good to very good model fit (IA > 0.5 generally considered good model fit, see chapter 3). The NMSE ranges from 0.053 to 0.354. FB ranges from -0.488 to 0.148. Weekly results show a wide range of R^2 results from 0.283 and 0.948 and results for IA range from 0.549 to 0.930, indicating good to very good model performance. NMSE differs between 0.031 and 0.306 and FB shows results from -0.487 to 0.158. For all levels of temporal aggregation the number of generally underpredicted stations as indicated by the FB is nine and the number of overpredicted stations is four. The development sites show some of the highest R^2 and IA results, indicating that the model also performs well at background locations. The NMSE at development site 13 however shows the highest NMSE for all levels of temporal aggregation and one of the highest underpredictions.

	ID	Hourly				Daily				Weekly			
		R^2	IA	NMSE	FB	R^2	IA	NMSE	FB	R^2	IA	NMSE	FB
Prediction Sites	1	0.434	0.784	0.244	0.101	0.536	0.793	0.161	0.100	0.562	0.713	0.080	0.115
	2	0.624	0.872	0.208	0.132	0.710	0.878	0.109	0.133	0.679	0.856	0.059	0.128
	3	0.713	0.867	0.300	-0.171	0.887	0.931	0.092	-0.172	0.886	0.895	0.067	-0.183
	4	0.740	0.877	0.286	-0.192	0.884	0.930	0.092	-0.181	0.892	0.904	0.054	-0.181
	5	0.332	0.715	0.481	-0.287	0.396	0.720	0.235	-0.288	0.304	0.549	0.152	-0.284
	6	0.754	0.794	0.395	-0.474	0.852	0.758	0.307	-0.488	0.691	0.603	0.306	-0.487
	7	0.470	0.770	0.311	0.150	0.528	0.741	0.201	0.148	0.283	0.552	0.136	0.158
	8	0.758	0.828	0.372	-0.368	0.920	0.851	0.198	-0.370	0.948	0.762	0.164	-0.372
	9	0.775	0.926	0.126	0.091	0.888	0.950	0.053	0.093	0.862	0.930	0.031	0.092
Development Sites	10	0.802	0.914	0.159	-0.254	0.888	0.917	0.107	-0.260	0.929	0.871	0.077	-0.251
	11	0.781	0.876	0.246	-0.293	0.898	0.888	0.152	-0.286	0.924	0.836	0.104	-0.286
	12	0.846	0.912	0.158	-0.199	0.946	0.928	0.080	-0.200	0.861	0.878	0.063	-0.204
	13	0.734	0.753	0.648	-0.469	0.909	0.779	0.354	-0.469	0.883	0.681	0.293	-0.476

Table 42: Model performance parameters (R^2 , IA, NMSE, and FB) for hourly, daily and weekly predictions of $PM_{2.5}$ concentrations by the LUR-adj model for monitored concentrations of 2010 at London ‘prediction’ (roadside monitoring stations) and ‘development’ (background monitoring stations) sites (using leave-one-out-cross-validation (LOOCV) to predict at ‘development sites’).

Summary:

The hourly LUR-adj predictions of $PM_{2.5}$ generally show a good linear relationship with R^2 above 0.5 for 10 out of 13 stations, the IA results are high (above 0.7) for all stations indicating a good model fit. Both NMSE and FB vary substantially between stations and suggest differences both in the extent of error ($0.119 < NMSE < 0.672$), as well as for the direction of over- or underprediction ($-0.474 < FB > 0.132$). Daily predictions show in comparison to hourly predictions slightly better results for model performance parameters R^2 , IA, and NMSE. Weekly model performance for R^2 is with 11 out 13

stations above 0.5 good. Some of the stations however show for weekly concentrations lower IA results compared to hourly or daily concentrations (e.g. station 5 and station 7). For these stations short-term (hourly and daily) variability may have been of particular importance and predicted variability from weekly averages (see Figure 31) may therefore not reflect monitored concentrations well.

Generally model performance is considered good; however a few stations perform less well for some model parameters than others. Two of these stations are station 5 and 7. These two stations had already shown particularly low agreement with other stations in step 1, above. A third station with comparably weak performance results is station 1, Brent - IKEA (see Figure 33), which shows comparable performance to other stations for IA, NMSE and FB, but some of the lowest results for R2 (hourly: 0.434, daily: 0.536, weekly: 0.562). A picture of station 1 is shown in Figure 33. It shows that the station is located next to a very busy road (London North Circular) with four lanes. The station is with about three metres from the kerb located comparably close to direct traffic influence. It is likely that the high source activity from the road could have caused variability in PM concentrations that is not well reflected by the predictions (resulting in a low R²).

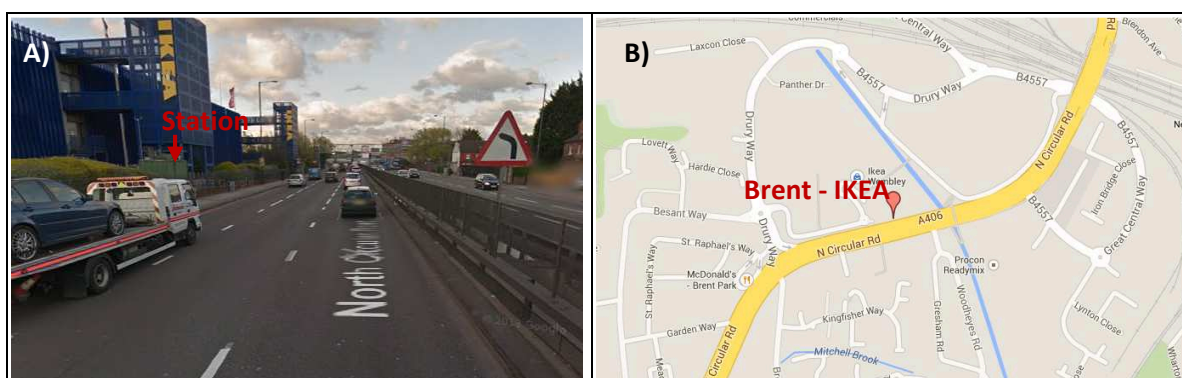


Figure 33: Street view (A) and map (B) of network monitoring station: Brent – IKEA (station ID 1) with locations of the station indicated. Source: Google Maps (<https://maps.google.co.uk>, accessed: 24th April 2014).

The performance parameters calculated for the development sites using LOOCV show at least similar (and for some parameters better) results compared to the prediction sites, confirming that for PM_{2.5} the model predicts also well at background monitoring stations.

LUR-adj predictions for PM₁₀

PM₁₀ concentrations at 26 stations have been modelled both by the LUR-adj and by the ADMS-urban models. Application of the ADMS-urban model was limited to the PM₁₀ prediction, as the model

does not predict $PM_{2.5}$. Results for model performance parameters for hourly predictions of the LUR-adj model and ADMS-urban model are presented in Table 43, better performance parameter results between the two models (by more than 0.01) are coloured in blue.

For hourly predictions with the LUR-adj model, the R^2 show a wide range from 0.246 to 0.780; IA are all above 0.5, and show differences between 0.608 and 0.938. Variability between stations can be observed for the error parameters of the LUR-adj model with NMSE varying between 0.064 and 0.516, and FB ranging from -0.550 to 0.140. In comparison to the LUR-adj model, hourly results for ADMS-urban predictions show a lower R^2 for all 26 stations ($0.080 < R^2 < 0.611$). IA results are also lower (by more than 0.01) for all but two stations; still all IA results are above 0.5, indicating good model fit. The NMSE is at 22 stations higher for ADMS-urban than for LUR-adj predictions, at only four stations the NMSE is lower for ADMS-urban. NMSE results for ADMS-urban range from 0.145 to 0.647.

For daily predictions, the LUR-adj model shows medium low to high R^2 results between 0.341 and 0.897. For ADMS-urban R^2 results lie in comparison between 0.082 and 0.879. For 22 out of 26 stations the R^2 results are higher for the LUR-adj model (by > 0.01). The LUR-adj model also shows better IA results (by > 0.01) compared to the ADMS-urban model for 20 out of 26 stations. IA results range from 0.612 to 0.971 for the LUR-adj model and from 0.531 to 0.955 for the ADMS-urban model indicating good model fit for all predictions. NMSE results are lower (by > 0.01) for most LUR-adj predictions compared to ADMS-urban predictions for 17 out of 26 stations. NMSE ranges between 0.015 and 0.377 for LUR-adj and between 0.026 and 0.428 for ADMS-urban. FB results are closer to 0 for LUR-adj compared to ADMS-urban predictions at 18 out of 26 stations (by > 0.01). Results for FB range from -0.550 to 0.139 for the LUR-adj model and from -0.408 to 0.030 for the ADMS-urban model.

ID	Hourly								Daily								Weekly							
	R ²		IA		NMSE		FB		R ²		IA		NMSE		FB		R ²		IA		NMSE		FB	
	LUR-adj.	ADMS	LUR-adj.	ADMS	LUR-adj.	ADMS	LUR-adj.	ADMS	LUR-adj.	ADMS	LUR-adj.	ADMS	LUR-adj.	ADMS	LUR-adj.	ADMS	LUR-adj.	ADMS	LUR-adj.	ADMS	LUR-adj.	ADMS	LUR-adj.	ADMS
1	0.712	0.549	0.728	0.727	0.369	0.271	-0.478	-0.330	0.854	0.771	0.672	0.760	0.283	0.156	-0.478	-0.330	0.728	0.700	0.506	0.627	0.273	0.134	-0.480	-0.334
2	0.523	0.492	0.818	0.698	0.176	0.313	-0.132	-0.317	0.716	0.663	0.880	0.733	0.076	0.196	-0.139	-0.324	0.674	0.571	0.804	0.601	0.050	0.157	-0.140	-0.327
3	0.727	0.542	0.918	0.769	0.086	0.227	0.071	-0.241	0.841	0.772	0.942	0.836	0.031	0.097	0.070	-0.242	0.746	0.688	0.904	0.703	0.017	0.079	0.080	-0.231
4	0.455	0.400	0.800	0.734	0.232	0.258	0.133	-0.069	0.724	0.720	0.892	0.900	0.066	0.054	0.132	-0.071	0.765	0.717	0.864	0.899	0.033	0.024	0.130	-0.069
5	0.508	0.371	0.777	0.692	0.206	0.255	-0.277	-0.297	0.665	0.620	0.769	0.726	0.116	0.137	-0.277	-0.297	0.559	0.589	0.599	0.595	0.098	0.113	-0.270	-0.288
6	0.246	0.112	0.608	0.500	0.502	0.647	-0.303	-0.355	0.429	0.217	0.692	0.579	0.195	0.284	-0.304	-0.356	0.368	0.159	0.551	0.460	0.120	0.176	-0.300	-0.337
7	0.780	0.591	0.938	0.804	0.064	0.178	0.024	-0.163	0.890	0.791	0.971	0.873	0.015	0.070	0.021	-0.165	0.848	0.744	0.952	0.806	0.010	0.055	0.010	-0.169
8	0.444	0.305	0.715	0.612	0.408	0.481	-0.301	-0.297	0.606	0.519	0.762	0.723	0.176	0.191	-0.296	-0.291	0.418	0.291	0.617	0.594	0.133	0.141	-0.290	-0.286
9	0.762	0.611	0.930	0.845	0.078	0.145	0.037	-0.064	0.897	0.879	0.971	0.955	0.017	0.026	0.033	-0.067	0.882	0.895	0.963	0.955	0.008	0.012	0.030	-0.065
10	0.632	0.489	0.822	0.676	0.233	0.413	-0.207	-0.323	0.867	0.806	0.889	0.782	0.072	0.158	-0.207	-0.322	0.865	0.821	0.801	0.683	0.053	0.118	-0.210	-0.319
11	0.408	0.360	0.743	0.661	0.294	0.336	-0.223	-0.251	0.497	0.584	0.770	0.750	0.127	0.125	-0.223	-0.252	0.458	0.510	0.637	0.616	0.073	0.079	-0.220	-0.243
12	0.704	0.603	0.890	0.809	0.161	0.255	0.059	-0.053	0.341	0.354	0.652	0.625	0.376	0.428	0.029	-0.084	0.320	0.377	0.641	0.675	0.132	0.143	0.020	-0.090
13	0.637	0.520	0.880	0.776	0.152	0.252	-0.021	-0.134	0.814	0.769	0.945	0.893	0.036	0.071	-0.021	-0.134	0.783	0.780	0.934	0.881	0.016	0.034	-0.020	-0.126
14	0.701	0.522	0.890	0.797	0.093	0.156	-0.143	-0.196	0.801	0.721	0.904	0.849	0.049	0.078	-0.142	-0.196	0.586	0.611	0.795	0.754	0.040	0.055	-0.130	-0.183
15	0.720	0.431	0.906	0.759	0.086	0.195	0.078	-0.201	0.830	0.606	0.928	0.809	0.035	0.095	0.076	-0.203	0.787	0.553	0.884	0.715	0.019	0.064	0.080	-0.192
16	0.628	0.413	0.881	0.656	0.108	0.362	-0.064	-0.414	0.725	0.581	0.897	0.643	0.048	0.236	-0.058	-0.408	0.578	0.514	0.827	0.473	0.024	0.193	-0.040	-0.385
17	0.561	0.432	0.856	0.780	0.173	0.218	0.076	0.027	0.716	0.599	0.911	0.875	0.054	0.070	0.079	0.030	0.568	0.418	0.845	0.799	0.034	0.046	0.090	0.042
18	0.674	0.560	0.892	0.805	0.114	0.175	0.111	-0.162	0.799	0.810	0.917	0.898	0.047	0.055	0.112	-0.159	0.797	0.867	0.887	0.876	0.025	0.032	0.110	-0.154
19	0.049	0.034	0.222	0.166	3.400	3.738	-0.143	-0.210	0.229	0.197	0.610	0.547	0.323	0.379	-0.145	-0.212	0.220	0.193	0.657	0.627	0.096	0.121	-0.130	-0.194
19b	0.319	0.229	0.689	0.569	0.439	0.546	-0.096	-0.163	0.568	0.509	0.849	0.784	0.089	0.120	-0.100	-0.167	0.450	0.416	0.797	0.754	0.044	0.060	-0.090	-0.154
20	0.647	0.523	0.771	0.837	0.297	0.145	-0.371	-0.015	0.806	0.750	0.756	0.929	0.191	0.039	-0.368	-0.012	0.706	0.667	0.599	0.899	0.158	0.019	-0.360	-0.007
21	0.620	0.501	0.876	0.702	0.118	0.309	-0.124	-0.382	0.711	0.674	0.903	0.716	0.049	0.188	-0.115	-0.372	0.488	0.401	0.810	0.561	0.031	0.148	-0.090	-0.345
22	0.709	0.558	0.900	0.837	0.116	0.161	0.140	-0.035	0.891	0.818	0.938	0.948	0.038	0.031	0.139	-0.035	0.877	0.809	0.891	0.943	0.026	0.014	0.140	-0.032
23	0.687	0.474	0.784	0.736	0.295	0.279	-0.345	-0.225	0.818	0.673	0.765	0.806	0.187	0.127	-0.347	-0.227	0.705	0.589	0.594	0.719	0.146	0.079	-0.350	-0.227
24	0.435	0.285	0.803	0.649	0.158	0.325	-0.022	-0.345	0.534	0.422	0.833	0.660	0.068	0.197	-0.020	-0.343	0.404	0.351	0.776	0.547	0.038	0.174	-0.030	-0.346
25	0.648	0.521	0.674	0.711	0.516	0.300	-0.550	-0.335	0.806	0.776	0.612	0.752	0.377	0.153	-0.550	-0.334	0.779	0.760	0.466	0.621	0.344	0.125	-0.550	-0.330
26	0.374	0.080	0.767	0.538	0.217	0.450	-0.119	-0.298	0.395	0.082	0.776	0.531	0.114	0.278	-0.119	-0.298	0.238	0.011	0.665	0.417	0.056	0.165	-0.110	-0.280

Table 43: Model performance parameters (R², IA, NMSE, and FB) for hourly, daily and weekly predictions of PM₁₀ concentrations by the LUR-adj model for monitored concentrations of 2010 data at ‘prediction sites’ (roadside monitoring station) in London. Better performance parameter results (by more than 0.01) between the two models are coloured in blue and bold.

For weekly concentrations R^2 results of the LUR-adj model are higher (by > 0.01) at 19 stations compared to ADMS-urban predictions. R^2 Results range from 0.238 to 0.882 for LUR-adj predictions compared to from 0.011 to 0.895 for ADMS-urban predictions. IA results are as well higher for LUR-adj predictions compared to ADMS-urban predictions at 17 out of 26 stations. Results for IA range between 0.466 to 0.963 for LUR-adj predictions and from 0.417 to 0.955 for ADMS-urban predictions. These results indicate that two stations show less than good model fit with $IA < 0.5$ for predictions of the LUR-adj model. For ADMS-urban prediction, model fit is below 0.5 at three stations. NMSE results range from 0.008 to 0.344 for LUR-adj and from 0.012 to 0.193 for ADMS-urban. The NMSE is for 17 out of 26 stations lower for the LUR-adj model compared to the ADMS-urban model. FB results range from -0.550 to 0.140 for LUR-adj predictions and from -0.385 to 0.042 for ADMS-urban predictions. For all levels of temporal aggregation, nine stations were over- and 17 stations underpredicted for the LUR-adj model. The ADMS-urban estimates underpredict for all stations apart from station number 17, which is narrowly overpredicted by the model.

Model performance results for LUR-adj predictions for the development site dataset (background locations) are presented in Table 44. For hourly concentrations R^2 results have a wide range between 0.160 and 0.818, IA results are between 0.454 and 0.938 with all but one station showing good model fit ($IA > 0.5$). Results for NMSE range from 0.070 to 0.976 and FB results range from -0.332 to 0.139. FB results indicate that concentrations at eight stations were generally underpredicted and concentrations at two stations overpredicted. For daily concentrations all performance results for R^2 , IA, and NMSE improved slightly compared to results for hourly concentrations ($0.444 < R^2 < 0.934$; $0.685 < IA < 0.974$; $0.016 < NMSE < 0.240$). FB results for daily concentrations range from -0.329 to 0.141. As for hourly concentrations, FB results indicate that concentrations at eight stations were over- and two stations underpredicted. For weekly concentrations, R^2 results show a wide range from 0.266 to 0.925. IA results however show good model fit for all stations between 0.556 and 0.965. NMSE results lie between 0.008 and 0.158 and FB results are between -0.320 and 0.140. FB results indicate that for weekly concentrations seven stations were underpredicted and one station overpredicted. FB at two stations is 0.000 indicating that the model neither over- nor underpredicts.

The results for the development sites generally show similar model performance compared to the prediction sites described above. Results for one station however (ID 30, City of London - Sir John Cass School) shows weak performance with for example results for hourly concentrations at: $R^2 = 0.160$, $IA = 0.454$, $NMSE = 0.976$, and $FB = -0.332$. Performance for daily and weekly concentrations are better, yet still weak in compared to other stations.

ID	Hourly				Daily				Weekly			
	R ²	IA	NMSE	FB	R ²	IA	NMSE	FB	R ²	IA	NMSE	FB
27	0.758	0.929	0.079	0.002	0.909	0.974	0.016	0.004	0.886	0.965	0.008	0.000
28	0.616	0.872	0.139	0.139	0.696	0.887	0.072	0.141	0.474	0.757	0.075	0.140
29	0.779	0.924	0.088	-0.077	0.896	0.953	0.031	-0.078	0.853	0.912	0.020	-0.070
30	0.160	0.454	0.976	-0.332	0.444	0.685	0.240	-0.329	0.266	0.556	0.158	-0.310
31	0.616	0.865	0.175	-0.007	0.830	0.945	0.037	-0.005	0.718	0.915	0.019	0.000
32	0.486	0.744	0.419	-0.206	0.715	0.830	0.124	-0.199	0.642	0.736	0.081	-0.190
33	0.738	0.924	0.084	-0.004	0.894	0.972	0.016	-0.005	0.844	0.957	0.009	-0.010
34	0.803	0.938	0.070	-0.058	0.934	0.973	0.018	-0.055	0.925	0.958	0.010	-0.050
35	0.818	0.900	0.119	-0.202	0.922	0.905	0.066	-0.201	0.903	0.813	0.052	-0.200
36	0.791	0.845	0.181	-0.313	0.897	0.817	0.129	-0.317	0.869	0.665	0.115	-0.320

Table 44: Model performance parameters for PM₁₀ of LUR-adj predictions of hourly, daily, and weekly monitored concentrations for 2010 at ‘development sites’ (background monitoring stations) in London, calculated using LOOCV (leave-one-out-cross-validation).

Summary

For the estimates at prediction sites, performance for most stations is good both for the LUR-adj and ADMS-urban predictions with IA indicating good model fit (above 0.5) for all stations for hourly and daily concentrations and for most stations for weekly concentrations. Two main observations can be made. The first observation is that for most stations the LUR-adj predictions perform better than the ADMS-urban predictions both for R² and IA.

The second observation is that performance clearly varies between stations. Between different stations the performance parameters by both models generally have a similar ranking. For example both show some of the weakest performance parameters for station 6 and some of the best results for station 9. This suggests a general ranking of the locations in terms of how easy or difficult they are to predict even with completely different methods. This is most likely related to strong local variability at some locations.

An example for one of the lowest performance results is station 6, City of London – Upper Thames Street. This station had shown weak correlations with other stations in step 1 (see Table 36). A picture of the site (see Figure 30) revealed its special location under a bridge, which may make it different and more difficult to predict compared to other locations. The second example of a station with comparably weak performance is station 26, Westminster – Marylebone Road (Figure 34, B). Particle concentrations monitored at Marylebone Road station have been identified by Beevers et al. (2013) as some of the highest concentrations at any London monitoring station. Reasons are that the station is located directly at the kerb of one of the busiest roads in central London with six different bus routes passing on the bus lane just next to the station.

A third station with comparably weak performance results is station 19b, for which outliers were removed earlier during data preparation (see above). The R^2 for hourly concentrations however (even after outliers were removed) are low with 0.319 for LUR-adj and 0.229 for ADMS-urban, pointing at a poor reflection of observed hourly variability by the models. The location as shown in Figure 34-B, while clearly at a major road, shows otherwise few specific characteristics in the immediate environment. A traffic sign next to the station however states “Queues likely” for the road, and frequent traffic jams with increased local source activity might therefore have been a potential cause for occasional short-term increases of concentration, resulting in increased local variability.



Figure 34: Street view of A) Westminster – Marylebone Road (Station ID 26), and B) Newham - Cam Road (station ID 19b) with the location of each monitoring station indicated. Source: Google maps (<https://maps.google.co.uk>; accessed: 7th Feb. 2014)

The results for the background stations (development sites) show for eight out of 10 stations good performance with similar or higher results compared to LUR-adj predictions at the roadsides stations (prediction sites). For one location however much lower model performance parameter results were calculated than at any other station: City of London – Sir John Cass School (stations 30). Station 30 has previously shown comparably low correlation with other stations (see Table 36). Its location is in a courtyard situation in central London (see Figure 30). Both the proximity to major roads behind the buildings, and the specific courtyard situation may have contributed to a different variability at this monitoring station compared to other locations.

4.5. Discussion

In this chapter a short-term model for ambient air pollution was successfully built using LUR models developed as part of the ESCAPE project combined with systematically selected temporal

adjustments. The resulting adjusted model shows good performance for hourly, daily, and weekly $PM_{2.5}$ and PM_{10} predictions of concentrations at monitoring stations in London. For PM_{10} , performance results exceeded predictions of the ADMS-urban dispersion model at most locations.

The main findings can be summarised as follows:

- The majority of monitoring stations in London showed a better correlation with the concentration average of all (other) background stations, compared to correlations with the nearest background station.
- Adjustments by ratio or by difference from the long term mean performed equally well.
- Temporal adjustment A (using ratios derived directly from hourly background concentrations) predicted 2010 variability at London roadside stations better compared with adjustment B (combining recognised temporal patterns of concentration variability in a ratio using hour of day, day of week, and season)
- Both adjustments A and B showed good spatial transferability to locations outside London. Adjustment B showed in addition good temporal transferability for PM_{10} when applied to 2011 concentrations at London roadside stations.
- All correlations for adjustment B however only showed low correlation ($r < 0.5$) with concentrations at stations in London both for 2010 and 2011, as well as with 2010 concentrations outside London. This approach is therefore considered of limited usefulness as a temporal adjustment for LUR models.
- Both for $PM_{2.5}$ and for PM_{10} the temporally adjusted 'LUR-adj' model (using adjustment A) showed good results for model performance parameters when predicting concentrations at locations in London.
- For PM_{10} , predictions of the LUR-adj model generally performed better than estimates of the ADMS-urban dispersion model when compared for London roadside stations.

One limitation applies to the development of the ADMS-urban model. A suburban background station was included in the regional background estimate. It is likely that the concentrations at the location to some extent included particle pollution from urban source activities. This may have caused overprediction of the regional background in the model. Final ADMS-urban predictions however showed under-prediction for 25 out of 26 stations, a large overprediction of the regional background was therefore considered unlikely.

The developed LUR-adj model clearly predicts well at most locations tested in London. For a few locations and most notably background station 30 (City of London – Sir John Cass School), some performance parameters showed however relatively weak results. The comparison with results from

the ADMS-urban model showed that some locations generally appear to be more difficult to predict than others regardless which of the two models were applied. It is likely that these stations have a particularly high influence of local factors, such as local wind patterns or specific location characteristics, such as the location under a bridge, as for location 6 (City of London - Thames Link Road). It would be difficult to include any of these factors into a model, as they appear to be very location specific. It would however be interesting for future research to investigate effects of certain local characteristics of particle distribution, such as under bridges or flyovers.

It should be noted that of the LUR-adj model only the land-use regression part is modelled. The adjustment has been added directly from monitored concentrations and can therefore not qualify as a model, which is defined as a 'simplification of reality' (see chapter 2). The term 'model' is however used here for the combined LUR-adj model.

One limitation of using temporal adjustments directly calculated from background monitoring data is the lack of temporal transferability. Adjustments need calculating for each time period the model is applied for. Background particle concentrations for London can however be downloaded from the LAQN website (<http://www.londonair.org.uk/london/asp/datadownload.asp>, accessed on 02. 04. 2014) for all years since 1993, which provides a broad range of years for future applications.

Spatial transferability of the LUR-adj model has been shown in a small application. The application tests direct spatial transferability of the model using concentrations from London monitoring stations. Results show that LUR-adj applications using London monitoring data can be considered for other locations in England. It is however recommended that transferability should be tested for each specific study area before application.

For two reasons the LUR-adj model is not directly transferable for applications in areas far away from London. The first reason is that the ESCAPE-LUR model developed for the region of London and the Thames Valley is of limited spatial transferability (Eeftens, Beelen, de Hoogh, et al. 2012). The second reason is that the temporal adjustment used for the LUR-adj model in this chapter was based on monitored concentrations in London. PM concentrations measured in London are not representative for areas that are influenced by different regional background concentrations.

The concept of the developed temporal adjustment is however transferable. Long-term predictions for an area can be calculated based on a different local LUR model. For many areas in Europe for example ESCAPE-LUR models are available, which could be utilised for long-term predictions (Eeftens, Beelen, de Hoogh, et al. 2012). Background concentrations based on monitoring stations in the area could then be applied to provide temporal adjustments.

LUR-adj predictions are subsequently used to calculate exposure for outdoor MEs, as well as for ambient components for other MEs as part of the personal exposure model (chapter 5 and 6).

5. Modelling In-Traffic and Indoor Microenvironments

5.1. Introduction

In this chapter a set of ME models, adjustments, and estimates are prepared for integration into the personal exposure model. Exposure in MEs differs from the surrounding ambient concentration. Each commonly visited ME is therefore modified before it is entered into the personal exposure model. For this purpose models and adjustments for several of the included microenvironments are compared and selected from the existing literature. For some other MEs, adjustments have been calculated using monitoring data as part of this thesis.

Included MEs fall into two main categories: transport and indoor microenvironments. The first section of this chapter focuses on adjustments and estimates of in-transport microenvironments. A second part focuses on models for home indoor concentrations, for which several existing indoor models from the literature are applied and compared for the prediction of monitored indoor concentrations. A small section about exposure adjustments for indoor office MEs follows. Each of these sections includes an introduction and a discussion.

5.2. Traffic Microenvironments

5.2.1. Introduction

In this section adjustments and estimates for in-traffic MEs are introduced for the MEs of cycling, bus, car, and rail transport. For some of these MEs, such as cars, studies exist which have calculated adjustment ratios for ambient to in-car concentrations. For other MEs, such as cycling or bus, only a few existing studies provide information on ME adjustments. Adjustment ratios were therefore calculated for the bus and cycling MEs from data collected during the transport monitoring campaign (described in chapter 3). Little is known about exposure in railway transport. A few studies do provide monitored concentrations inside trains. Estimates for exposure in railway transport systems are selected from these existing studies and introduced at the end of this section.

The following paragraphs first describe how concentrations along routes are prepared using the ESCAPE-LUR model. Next, the selection or development of adjustments for different transport

microenvironments are presented, starting with cycling, followed by bus, car, and finally rail transport.

5.2.2. Data Preparation

Concentrations along routes were predicted using the temporally adjusted land-use regression (LUR) model, as described in chapter 4. Exposure in different road traffic modes can then be altered from these concentrations in a second step.

Modelling Routes for Personal Exposure

As a first step, regular grid points of 20m distance for the London area were intersected with a buffer around the central road line (road data source: Meridian). In Figure 35-A, a sample route part with intersected grid points is shown. A grid distance of 20m was chosen in order to achieve good representation of locations along the route. The buffer size of 20m around the road was chosen to approximately cover the road area widths (road plus sidewalk). For estimates of personal exposure the average concentrations predicted for all grid points along the route was calculated.

Preparing Monitored Concentrations for the Calculation of Transport ME Adjustments

For data collected during the transport monitoring campaign two transport modes were monitored simultaneously along the routes. A detailed comparison between the two modes for segments passed along the route was intended as preparation for the calculation of in-transport adjustments. As a preparation, monitored minute concentrations along the route were attached to each GPS point according to time stamp (up to four GPS points with (intervals of up to 15sec) have the same minute concentration attached). Monitored concentrations along the route were then aggregated for small road segments for each repeated journey. 20m segments were chosen for the initial data aggregation in order to A) reflect high spatial accuracy, and B) get an average of several GPS points for every segment. GPS points were then averaged for 20m buffers around each segment. Using concentrations by segments a direct comparison could be made between transport modes along the routes. The buffers overlap and the average therefore effectively smoothed data in a 60m sliding average along the route (see Figure 35-B). This widens the data base used for averaging concentrations per segment, and can smooth typical short-term traffic related concentration peaks (e.g. single passing car) along the road.

For the calculation of the adjustment ratios the concentrations were averaged for the whole route.

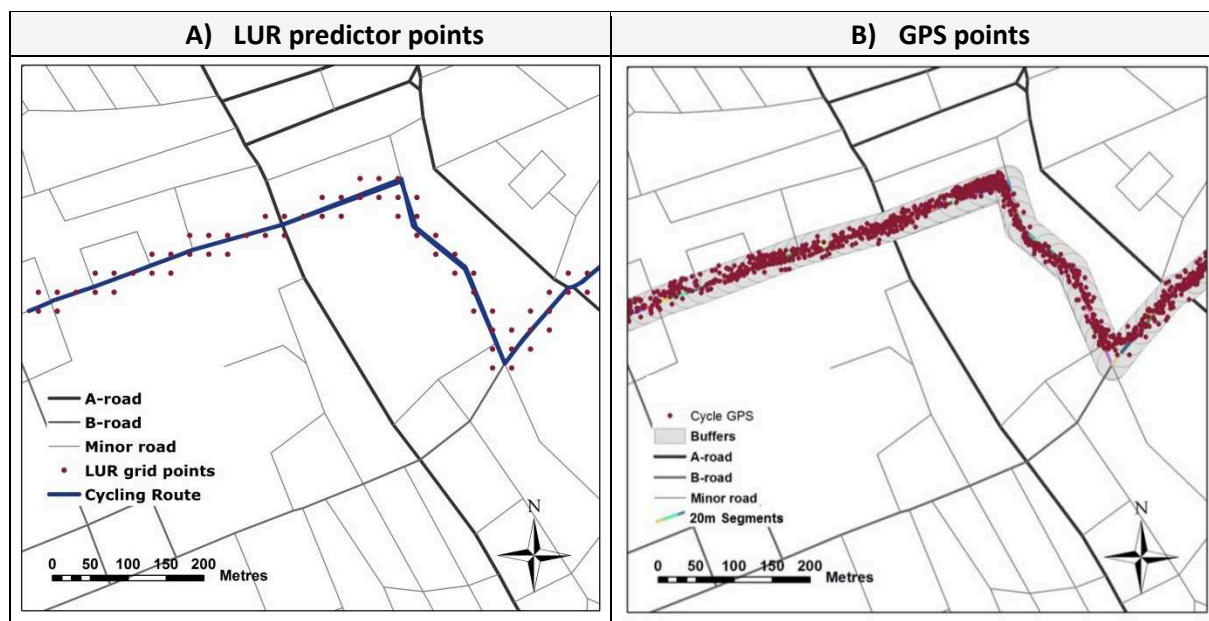


Figure 35: Route section from the cycling commute of individual A, with A) points of LUR model receptors, and B) GPS recorded points (including 25 repeats) with 20m buffers. Road data source: Meridian

5.2.3. Adjusting for Cycling Exposure

It has been shown before (chapter 2) that particles disperse rapidly away from the kerb and exposure on the sidewalk is generally lower compared to direct in-traffic exposure (Kaur et al. 2005). Road estimates by the LUR model effectively predict concentrations on the sidewalk, as the monitors underlying the model are located several meters from the kerb. In central London cyclists generally share the main traffic lanes with other vehicles and therefore experience direct exposure to vehicle sources. To account for the horizontal displacement between the LUR roadside estimates and cyclists' in-traffic exposure adjustments were undertaken in form of simple ratios. The ratios were calculated from simultaneously undertaken walking and cycling concentrations monitored along the same routes. Not adjusting for this difference could lead to underestimation of in-traffic exposure and to exposure misclassification. In addition, cycling to background ratios and walking to background ratios were calculated from simultaneously monitored concentrations at a background site (see chapter 3). For the cycling to background ratios concentrations collected during the cycling-bus (see chapter 3) and the cycling-walking campaigns were pooled in order to acquire a bigger data base of altogether 18 journeys for the calculation of the ratio each for the low traffic and high traffic route.

Results for cycling to walking, as well as cycling to background and walking to background ratios are presented in Table 45. All ratios are above one, indicating generally higher exposure for the cyclist compared to walking or concentrations at the background location. For PM_{2.5} the mean cycling to

walking ratio along the low traffic route is with 1.39, a little below the mean ratio along the high traffic route with a ratio of 1.68. Similarly, for PM₁₀, the ratio for the low traffic route lies below the ratio on the high traffic route with 1.21 and 1.36 respectively. This difference most likely reflects a steeper gradient for particle dispersion between concentrations close to traffic for high traffic routes compared to low traffic routes. SD for all cycling to walking ratios is moderate with all SD below 1 (both PM_{2.5} and PM₁₀). The range is for PM_{2.5} is however high with a range of more than twice the mean ratio for both route types.

		Mean	SD	Range
PM _{2.5}	Cycle-walk BG	1.39	0.66	3.60
	Cycle-walk TR	1.68	0.80	4.12
	Cycle-background BG	1.57	0.44	1.90
	Cycle-background TR	2.01	0.50	1.89
	Walk-background BG	1.22	0.50	1.49
	Walk-background TR	1.33	0.36	0.96
	Walk-background both	1.27	0.43	1.49
PM ₁₀	Cycle-walk BG	1.21	0.42	1.23
	Cycle-walk TR	1.36	0.36	0.98
	Cycle-background BG	2.21	0.98	3.46
	Cycle-background TR	2.41	0.82	2.84
	Walk-background BG	2.05	0.80	2.38
	Walk-background TR	2.24	0.36	0.95
	Walk-background both	2.13	0.63	2.38

Table 45: Cycling to walking and cycling to background ratios for low traffic (BG) and high traffic (TR) routes, both for PM_{2.5} and PM₁₀.

The cycling to background ratios are all higher than the cycling to walking ratios for respective route type and particle size fraction. As for the cycling to walking ratios, the cycling to background station ratios are lower for the low traffic routes compared to the high traffic routes. For PM_{2.5} the mean ratios are 1.57 for the low traffic and 2.01 for the high traffic route. For PM₁₀, mean ratios are 2.21 for the low traffic and 2.41 for the high traffic route. The SD for cycling to background ratios are all below 1 (both for PM_{2.5} and PM₁₀), the range of cycling to background ratios is also moderate with less than twice the mean.

Walking to background ratios are lower compared to cycling to background ratios within each route type and size fraction. In addition, the high traffic route shows higher walking to background ratios compared to the low traffic route for both particle size fractions. Mean ratios for PM_{2.5} are 1.22 for the low traffic route and 1.33 for the high traffic route. SD results are comparably low with 0.50 for the low traffic route and 0.36 for the high traffic route. For PM₁₀, the mean ratio for the low traffic route is 2.05 and the mean ratio for the high traffic route is 2.24. SD is, with 0.80 (low traffic route)

and 0.30 (high traffic route), comparably moderate. The combined walking to background ratios for both high traffic and low traffic are also presented here, as a combination of low traffic and high traffic routes most likely represents the mixed purpose and attitude towards walking journeys. Some walking journeys are undertaken as commutes (much less than for all other transport modes) when people might choose to follow more direct routes along highly frequented traffic corridors. Other journeys are however undertaken for leisure (more than for other transport modes), when people would have time to choose longer routes to follow less heavily trafficked roads (Transport for London 2008a; 2008b). For $PM_{2.5}$ the mean ratio for both routes combined is 1.27 (SD: 0.43, range: 1.49), for PM_{10} the mean ratio is 2.13 (SD: 0.63, range: 2.38).

Variability along the route

The dataset collected for simultaneous walking and cycling during the campaign was small with only 10 repeats along the low traffic route and 8 repeats along the high traffic route. For small data samples the influence of each data point is increased, a single outlier could for example cause a substantial change to the mean concentration. In order to assure that the results are representative for cycling to walking ratios, the variability of ratios along the route, as well as between repeated journeys was examined. Parameters for data variability, such as standard deviation are also less stable for small datasets; therefore the data was made visible in several graphs and interpreted directly. Visually one can detect unusual data points, as well as potential patterns in the variability.

Concentrations along the routes were summarised for 20m segments, as described above and then calculated into ratios. The spatial variability of cycling to walking ratio along the route is presented in Figure 36. The graph shows the average ratio for each continuous road segment from start (left) to finish (right) of each route. The graphs show that ratios vary considerably along both routes and for both particle size fractions. For several areas along the route high or low ratios prevail over several sequential segments. These patterns may reflect certain street configuration or traffic situation, such as distance of pedestrian from kerb, or width of road. The ratios along the route do not however show clear outliers, which may have affected the mean cycling to walking ratio.

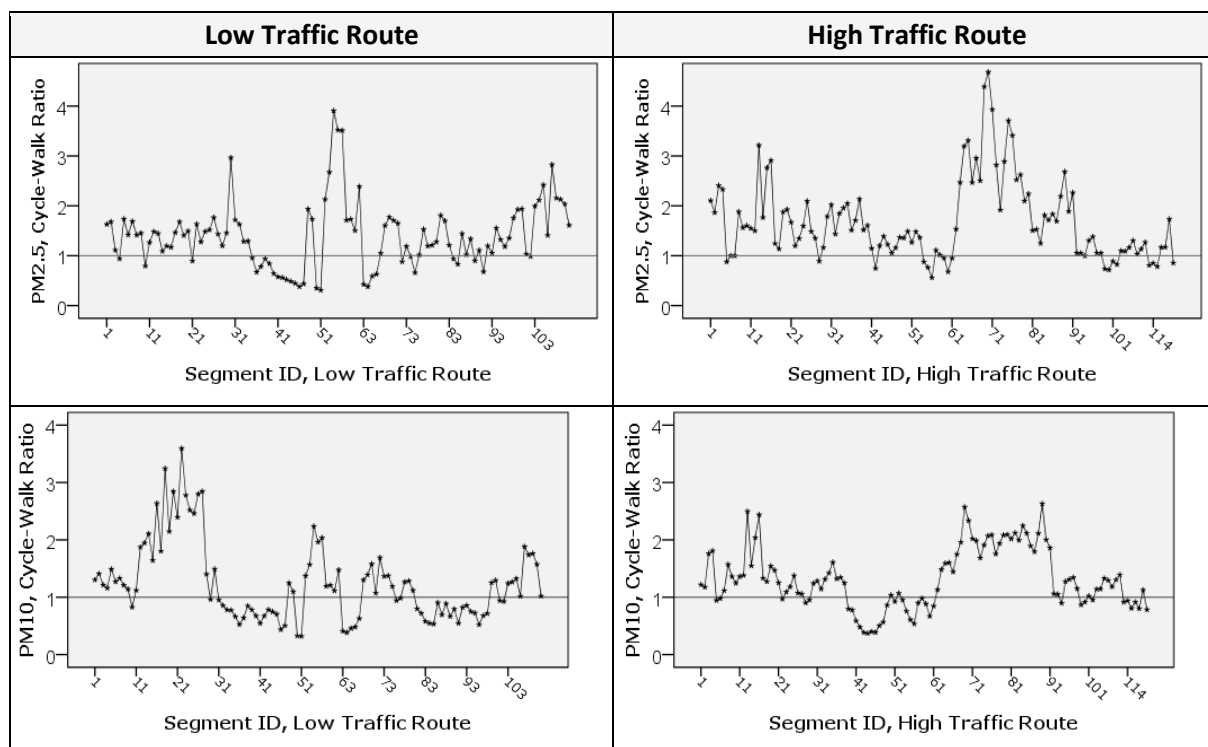


Figure 36: Cycling to walking ratio for PM_{2.5} and PM₁₀ averaged per road segment with continuous road segment numbers along the routes from start (left) to finish (right) for low and high traffic routes.

Variability between repeated journeys

The variation between repeated journeys (calculated from mean concentrations per segment) was examined in Figure 37. The repeat numbers in the graph are sequential throughout the monitoring campaign (10 days) and the same repeat number for the two routes refers to repeats undertaken during the same morning or afternoon of the same day.

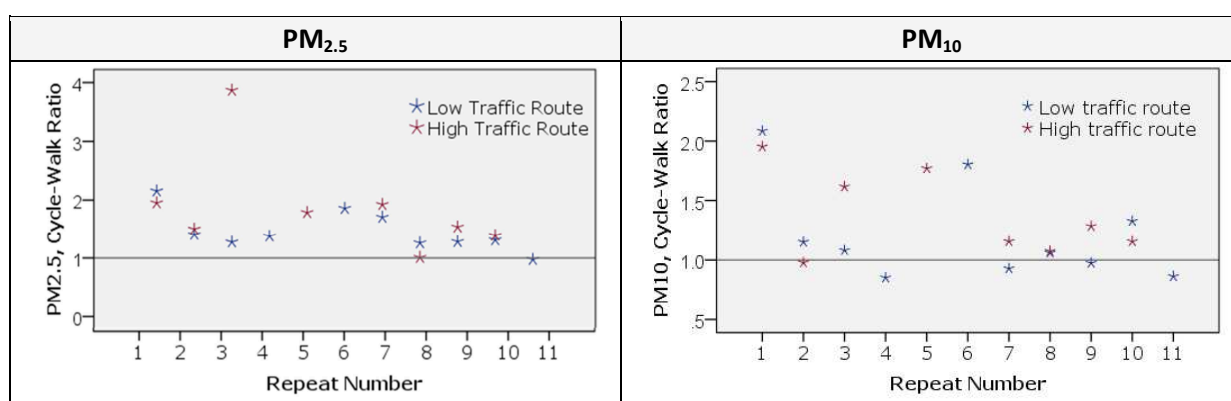


Figure 37: Cycling to walking ratios for PM_{2.5} and PM₁₀ by sequential number of repeat (repeats show gaps where monitoring during repeats failed) for high and low traffic routes.

Figure 37 shows that both for $PM_{2.5}$ and PM_{10} , ratios vary between different repeated journeys with data points for the majority of repeats (both routes) between 1 and 2. One ratio for $PM_{2.5}$ along the high traffic route however lies clearly above all other results, which would have affected the mean ratio. Differences between the repeats could have a multitude of reasons. Possible reasons could for example include differences in wind direction (wind blowing from the road to the sidewalk, lowering difference between modes) or different traffic experienced between the modes.

Conclusions

For $PM_{2.5}$ three existing studies also calculated results of simultaneously monitored walking and cycling concentrations along high traffic routes. One study by Kaur et al. (2005) measured a total of 48 repeats (repeated monitoring along the same route) for cycling and 56 repeats for walking along two different routes in central London. A study by McNabola et al. (2008) simultaneously monitored cycling and walking for two routes in Dublin. The number of repeats is given as 48 for cycling and 46 for walking (both routes together). A second study by McNabola et al. (2009b) monitored cycling and walking simultaneously along one circular route in Dublin, Ireland for 50 times. The cycling to walking ratios for the studies are 1.22 (calculated from mean concentrations per mode) by Kaur et al. (2005), 1.39 by McNabola et al. (2009a), as well as 1.41 and 1.75 for the two routes by McNabola et al. (2008). The mean ratio calculated as part of this thesis lies with 1.68 within the range of results from these studies.

Mean ratios for cycling to walking calculated for this thesis are based on a low number of repeats. In addition, for $PM_{2.5}$ one out of eight repeats for the high-traffic route showed a result more than double the average, which would have had a substantial influence on the mean ratio. For $PM_{2.5}$, several studies exist which also calculated cycling to walking ratio, based on a larger sample number. It was therefore decided that for $PM_{2.5}$ a ratio from the literature with more repeated journeys would provide a more representative estimate for an adjustment. Of the available studies, the study by Kaur et al. (2005) was considered less suitable, as different numbers of repeats for cycling and walking were given from which the cycling to walking ratio was then calculated. This implies that not all the repeats were with a simultaneous match from the other transport mode. Of the two studies by McNabola, the study from 2008 provides less journey repeats for each of the two routes (between 46 and 48 repeats when combined), compared to the study from 2009 (50 journey repeats). It was therefore decided to use the ratio by (McNabola et al. 2009a) of 1.39 to adjust $PM_{2.5}$ from LUR predictions to in-traffic exposure instead of monitoring results from this thesis. A limitation of the study by McNabola et al. (2009b) was however that monitoring took place during

very high traffic conditions. Results acquired for this specific situation may not reflect cycling on routes with less traffic well. The study by McNabola for 2008 showed similarly high traffic. Using results from McNabola et al. (2008) would therefore have had the same limitation.

For PM_{10} no other studies from the literature provided simultaneous walking and cycling monitoring data that could be considered for application. The ratio calculated from data collected during the mode comparison campaign was therefore used as an adjustment for sidewalk to cycling exposure. Most cycling journeys are undertaken during commutes (Transport for London 2008). Commutes by default take place during rush-hour conditions, it was therefore decided that the high traffic average would better reflect this traffic situations and is therefore used to adjust concentrations. The adjustment factor applied to LUR roadside predictions for PM_{10} of 1.32 was therefore used.

Ratios between cycling and background location have been calculated as well by Zuurbier et al. (2010). Zuurbier et al. (2010) measured $PM_{2.5}$ and PM_{10} during 16 cycling journeys along a high traffic and a low traffic route during rush hours in Arnhem (the Netherlands). Results were compared to a stationary background monitor. Results show ratios for $PM_{2.5}$ are 2.11 on both low and high traffic route. This ratio is markedly higher than the result for the low traffic route from this thesis with 1.37, the ratio for the high traffic route from this thesis is closer with 1.94. For PM_{10} the study by Zuurbier et al. (2010) shows on the other hand lower cycling to background ratios compared to results from this thesis with 1.10 on the low traffic and 1.13 on the high traffic route, compared to 1.65 on the low traffic and 2.35 on the high traffic route in this thesis. Differences to the results by Zuurbier et al. (2010) may to some extent be explained by differences between the study areas. It is likely that a cycling route in the Netherlands would for the most part follow dedicated cycling paths, in England however, mode sharing between cyclists and motorised vehicles is much more common. For studies in England the ratio calculated by this thesis is therefore be considered more representative.

One other study, Kaur et al. (2005a) calculated a walking to background ratio for $PM_{2.5}$. This study showed a ratio for walking to background location for major road in London (Marylebone Road) of 2.34. This ratio lies well above the ratios calculated in this thesis for both low traffic (1.22) and high traffic (1.33) route. The difference is most likely due to the specific location of the monitoring campaign along one of the busiest roads in central London.

5.2.4. Adjusting for In-Bus Exposure

In this section an adjustment from sidewalk to in-bus concentrations is developed by first calculating a ratio between in-bus and cycling exposure, and then combining the adjustment with the cycling to sidewalk ratio (developed above). In addition, in-bus to background ratios are calculated. In-bus

exposure has been shown by many studies to differ from concentrations along the sidewalk or at background locations (chapter 2). Several studies which compared different transport modes along the same route found that particle concentrations inside buses exceed concentrations in most other road traffic microenvironments. High in-bus concentrations have been connected on one hand to close proximity to traffic sources and on the other hand to additional in-bus sources, such as resuspension from people's movement. Many further influences on in-bus concentrations have been suggested in different studies and were described in chapter 2. The multitude of different influences on in-bus concentrations makes the development of a more sophisticated, yet transferable model for in-bus concentrations difficult. A simplistic approach has therefore been adopted here. For the calculation of the in-bus to cycling ratio, monitoring data from the transport monitoring campaign were used, for which concentrations were collected simultaneously for cycling and inside buses along two routes (see chapter 3). Repeat numbers for both routes were relatively low with 15 for low traffic and 17 for high traffic routes. As mentioned above, for low numbers of data points parameters such as the mean and SD can be unreliable. In addition to looking at mean, SD and range of the data, visualisation of the data variability was therefore undertaken and examined directly.

In Table 46 basic statistics for the in-bus to cycling and in-bus to background ratios are presented. For the in-bus to cycling ratios, mean $PM_{2.5}$ ratios are 0.91 for the low traffic and 1.06 for the high traffic route. SD is relatively small with 0.28 for the low traffic and a little higher with 0.38 for the high traffic route. The mean in-bus to cycling ratios is generally higher for PM_{10} than for $PM_{2.5}$. The PM_{10} ratios are 1.37 for the low traffic and 1.80 for the high traffic route. The SD is 0.43 for the low traffic and 0.78 for the high traffic route. As for $PM_{2.5}$, the high traffic route ratio shows a higher mean, as well as a higher SD, compared to the low traffic route.

		Mean	SD	Range
$PM_{2.5}$	Bus-cycling BG	0.98	0.28	1.02
	Bus-cycling TR	1.33	0.38	1.38
	Bus-background BG	1.50	0.55	1.69
	Bus-background TR	2.06	1.22	3.75
PM_{10}	Bus-cycling BG	1.49	0.43	1.26
	Bus-cycling TR	1.95	0.78	2.50
	Bus-background BG	2.89	1.19	3.70
	Bus-background TR	4.14	2.53	8.46

Table 46: In-bus to cycling and in-bus to background ratios for low traffic (BG) and high traffic (TR) routes, both for $PM_{2.5}$ and PM_{10} .

In-bus to background ratios generally lie above the in-bus to cycling ratios for the respective route type and particle size fraction, which reflects the higher distance of the background station to the

road source, compared to roadside measurements. Both for $PM_{2.5}$ and PM_{10} , the in-bus to background ratios are higher for the high traffic route compared to the low traffic route. For $PM_{2.5}$, the mean in-bus to background ratio is 1.50 for the low traffic route and 2.06 for the high traffic route. For PM_{10} the mean ratio is 2.89 on the low traffic and 4.14 on the high traffic route. SD for $PM_{2.5}$ are 0.55 and 1.22 for the low traffic route and the high traffic route respectively, for PM_{10} , the SD is 1.19 for the low traffic route and 2.53 for the high traffic route respectively.

In Figure 38 the mean in-bus to cycling ratio is presented for sequential segments from the start (left) to the finish (right) of the routes. The SD around the mean is shown with a multiplier of 2 (referred to as $SD*2$; this SPSS standard setting was used for better visualisation of the effect) as error bars around the mean. As presented in chapter 2, changes of in-bus to outdoor concentrations along their routes have been connected to bus idling, door openings and high traffic situations (Vijayan & Kumar 2010; Song et al. 2009). For a more complete picture of the bus routes, relevant situations for these potential influences have therefore been indicated in the graphs: road segments that include a bus stop (door opening, idling), traffic lights along the route (idling), major roundabout (increased outside traffic), and school bus stops (in-bus volunteer reported passenger numbers filled to full capacity during school rush hours). Mean of the segment averages, SD and range (of segment averages) are added to the graph.

The variability between road segment averages shows moderate SDs of 0.38 (low traffic route) and 0.56 (high traffic route) for $PM_{2.5}$. For PM_{10} , the SD are generally higher, with 0.64 for the low traffic and 0.83 for the high traffic route. The mean concentrations in the graphs (both $PM_{2.5}$ and PM_{10}) show mostly a continuous pattern of rising and falling ratios along the route for the low traffic route. In comparison, for the high traffic route the mean segment concentrations show more 'jumps' between low and high ratios for neighbouring segments. For the $SD*2$ between the segments large differences can be observed, in particular for the high traffic route. Increased traffic sources are a likely explanation for higher variability along the high traffic compared to the low traffic route.

An interpretation of events along the routes (bus stops, traffic lights, roundabout) is difficult from this small dataset. The high frequency of bus stop and traffic lights (high traffic route) make it additionally difficult to connect one situation with a specific segment. The major roundabout and the school bus stops coincide with added variability in form of increased $SD*2$ and increased mean ratios for (and just after) the affected segments. In reference to the findings from other studies these situations may have influenced the ratio. A broader evidence base would however be necessary to draw conclusions from these connections.

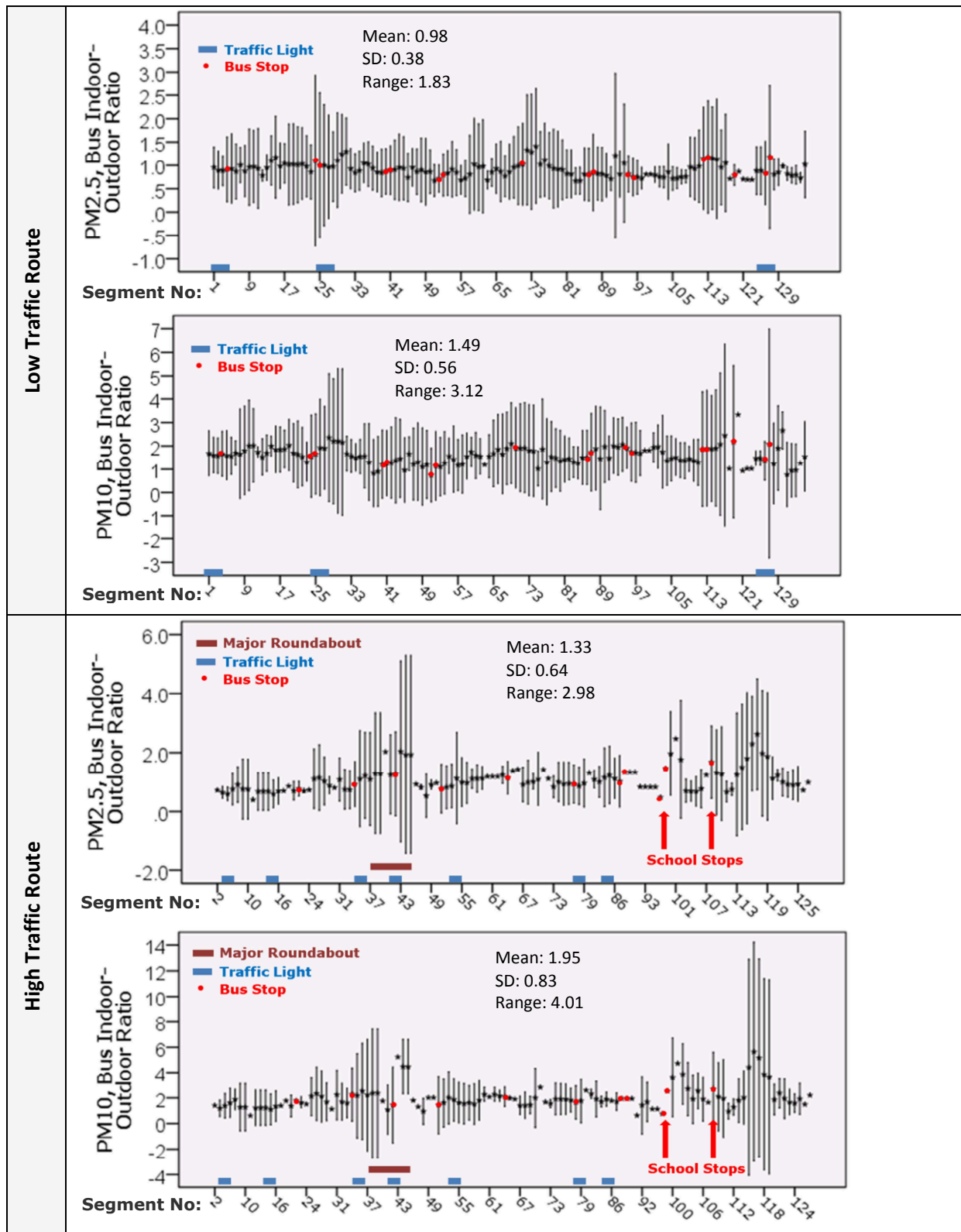


Figure 38: Mean PM_{2.5} and PM₁₀ in-bus/cycling ratios for continuous road segment numbers from start (left) to finish (right) along the low traffic and high traffic route, with variability displayed (2 * Standard Deviation) around the mean. Mean, SD (Standard Deviation), and range are added. Specific situations along the routes are indicated.

In Figure 39, variability of in-bus to cycling ratios between different journeys (repeats) is visualised per route. The same repeat number for low and high traffic routes refers to the same monitoring time period (same morning or afternoon of the same day). For $PM_{2.5}$ most ratios for different repeats lie between 0.5 and 1.5 both for the low traffic and the high traffic route. For PM_{10} most of the ratios for different repeats lie between 1 and 2.5. For both $PM_{2.5}$ and PM_{10} the graph does not show any clear outlier.

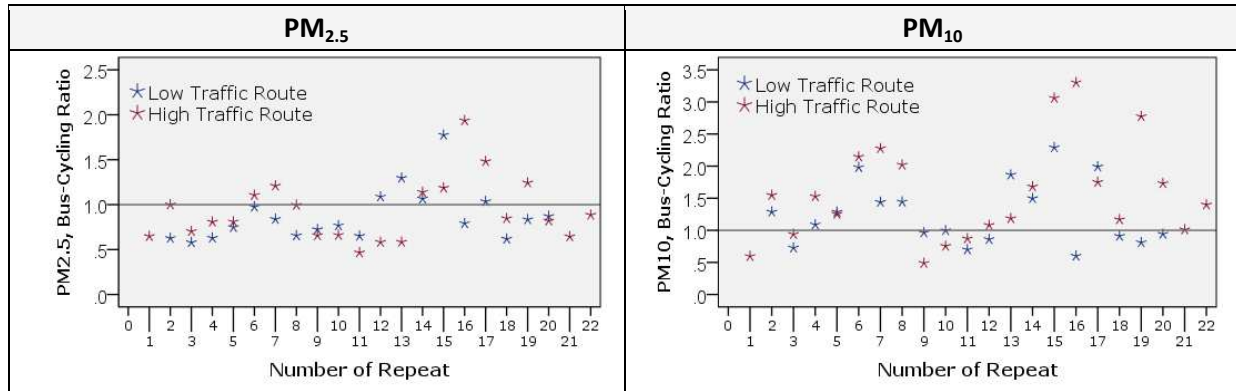


Figure 39: Average in-bus/cycling ratio for each sequential repeat number along the low traffic and high traffic routes for $PM_{2.5}$ and PM_{10} (Repeats show gaps when monitoring failed).

Calculating an In-Bus to Roadside Ratio

From the mean ratios between in-bus and cycling, as well as cycling to roadside, an in-bus to roadside ratio was calculated. Bus routes in most places follow along major roads. In order to calculate a representative in-bus to roadside adjustment, the high traffic route ratios are more representative of most bus routes and were therefore chosen for this calculation. The adjustment from the roadside to in-bus concentrations (PM_{bus}) is calculated by combining roadside to in-traffic adjustment, as presented above, with in-traffic to in-bus adjustment, which can be written as follows:

Equation 6

$$PM_{bus} = PM_{LUR} * (t/s) * (b/t)$$

With the predicted LUR modelled route estimates (PM_{LUR}) multiplied by the in-traffic to sidewalk ratio (t/s) and by the in-bus to in-traffic ratio (b/t). Adding the adjustments factors into the equation for $PM_{2.5}$ and PM_{10} , the resulting equations read as follows:

$$PM_{2.5_{bus}} = PM_{LUR} * 1.24 * 1.33 = PM_{LUR} * 1.65, \text{ and}$$

$$PM_{10_{bus}} = PM_{LUR} * 1.32 * 1.95 = PM_{LUR} * 2.57$$

Conclusions

In summary, in-bus to in-traffic ratios vary temporally as well as spatially with the mean ratios for $PM_{2.5}$ showing generally less variability compared to the mean PM_{10} ratios. For several repeats $PM_{2.5}$ ratios are below 1, indicating that on several journeys the in-bus exposure was below the cyclist's exposure. This is however contrary to findings from the literature, which show higher mean concentrations inside buses compared to exposure for cyclists (Zuurbier et al. 2010; McNabola et al. 2008; McNabola et al. 2009a).

For $PM_{2.5}$, ratios of in-bus to walking and in-bus to cycling have been calculated as well by McNabola et al. (2008), based on simultaneous measurements. Results for in-bus to walking ratios are 3.09 and 2.14 (different routes) and in-bus to cycling ratios are 1.38 and 3.21 (different routes), both collected along a high traffic route. Another study by McNabola et al. (2009) provides data for simultaneously monitored cycling, walking and in-bus $PM_{2.5}$ concentrations along one high traffic route. Ratios can be calculated from the results of 2.02 for in-bus to walking and of 1.45 for in-bus to cycling. Mean ratios along the traffic route for this thesis are 1.33 for in-bus to cycling and 1.65 for calculated in-bus to walking, which is generally lower compared to the findings by either of the studies by McNabola (2008; 2009). Both studies by McNabola have however been conducted during times of very high (congested) traffic, which most likely influenced results.

In-bus to background location ratios were calculated by several other existing studies. A study by Zuurbier et al. (2010) for Arnhem (Netherlands) calculated ratios of diesel bus to background concentrations from concentrations collected during 10 journeys undertaken during rush hour times. For $PM_{2.5}$ the calculated ratio by Zuurbier et al. (2010) is 2.98. A study by Asmi et al. (2009) collected in-bus $PM_{2.5}$ concentrations for 8 days for a study in Helsinki, Finland. Ratios between in-bus to background location were calculated and lie between 0.95 and 2.87 (for different days and bus ages). In-bus to background ratios for this study (low traffic route: 1.50, high traffic route: 2.06) are lower compared to results from Zuurbier et al. (2010), but lie in between the range of results from Asmi et al. (2009).

For PM_{10} , the study by Zuurbier et al. (2010) calculated a diesel bus to background ratio of 2.49. Praml & Schierl (2000) have reported in their study on public transport in Munich (Germany) that in-bus (and in-tram) concentrations for PM_{10} are 3 to 5 times the ambient concentration monitored at a nearly roadside station. In-bus to background ratios from this study (low traffic route: 2.89, high traffic route: 4.14) lie above the ratio calculated by Zuurbier et al. (2010). The ratio for the traffic route falls within the range of results suggested by Praml & Schierl (2000).

Results of the in-bus to cycling ratios calculated above show no apparent outliers for spatial or temporal variability. The ratios were therefore considered representative for the ratios between in-bus and cycling along the route. Existing studies by McNabola et al. (2008; 2009a) have collected in-bus and cycling $PM_{2.5}$ concentrations simultaneously along a route. Results from these studies have been collected during traffic congestion and are therefore potentially not representative for other less congested routes. For adjustment of roadside to in-bus concentrations as part of the personal exposure model, the ratios calculated above of 1.65 for $PM_{2.5}$ and 2.57 for PM_{10} have therefore been applied.

For ratios of in-bus to background concentrations two existing studies are both based on smaller sample sizes compared to results from this thesis. The in-bus to background ratios calculated for this thesis are therefore considered in comparison more representative.

5.2.5. Adjusting for In-Car Exposure

In this section an adjustment from ambient concentrations to exposure inside cars is selected from existing results in the literature. Exposure in cars has been studied more than other transport modes and a few studies have developed ambient to in-car adjustment ratios. Studies on in-car exposure have been introduced in chapter 2.

Studies presented in chapter 2 showed that exposure inside cars exceed concentrations at ambient monitoring stations. Ratios between in-car and background monitoring stations have been calculated by Zuurbier et al. (2010) $PM_{2.5}$ of 1.57 (Diesel car) and of 1.95 (Gasoline car), as well as for PM_{10} of 1.44 (Diesel car) and 1.26 (Gasoline car).

One study by McNabola et al. (2008) calculated in-car to walking ratios for $PM_{2.5}$ from simultaneously monitored concentrations. Ratios are 1.34 and 1.57 for different routes. As described before, the study by McNabola et al. (2008) was however conducted during times of congested traffic, which may affect the transferability of the ratio. For PM_{10} as well one study by Gulliver & Briggs (2007) provides in-car to pedestrian ratio based on simultaneous measurements. The study calculated an in-car to walking ratio of 1.42.

The limited number of existing studies on in-car to roadside ratios provides no scope for selection. The average of the two provided estimates by McNabola et al. (2008) of 1.46 is applied as adjustment for $PM_{2.5}$. The adjustment provided by Gulliver & Briggs (2007) of 1.42 is applied as adjustment for PM_{10} . Ratios from the presented studies for in-car to walking ratios are applied as adjustments from roadside to in-car concentrations for the personal exposure model.

5.2.6. Estimates for Exposure in Railway Transport

Another important mode of transport is railway transport. Different types of local railway transport exist, for example local trains (such as the London overground), subway trains or trams. Most local railway systems are separated from road transport and are influenced by different particle sources, such as breakware, as described in chapter 2. In addition, some rail routes may be under ground or in tunnels, such as parts of the London underground system. Railways are therefore extremely difficult to model or adjust from ambient concentrations.

Very few studies have been conducted on particle exposure in railway transport. For the personal exposure model, it was however considered important to include an estimate for times in railway transport, as it has been shown that exposure in railway transport is often higher than ambient concentrations. A few existing studies on railway transport have been conducted for the London underground. The highly increased concentrations during times in the tunnel were however estimated to be up to 10 times above concentrations when above ground (Hurley et al. 2004; Kam et al. 2011). The majority of people's railway journeys are however undertaken above ground (e.g. for London: more than 50% of the London underground are above ground, see also: http://en.wikipedia.org/wiki/London_Underground). Studies focusing on concentrations under ground were therefore not considered representative for general railway transport. A study by (Hurley et al. 2004) on London tube trains provided estimates for the difference between above and below ground sections of several different lines. From this study an estimate for railways above ground could as such be calculated (assuming 50% of times above and below ground). Calculated estimates for sections above ground are $12.12\mu\text{g}/\text{m}^3$ for the Central line, $26.04\mu\text{g}/\text{m}^3$ for the Northern line, and $26.04\mu\text{g}/\text{m}^3$ for the Victorian line. The average of these three estimates is $20.64\mu\text{g}/\text{m}^3$. In the absence of any more sophisticated method to estimate $\text{PM}_{2.5}$ concentrations for times in railway transport, the fixed concentration of $20.64\mu\text{g}/\text{m}^3$ was used to estimate $\text{PM}_{2.5}$ exposure during railway transport for the personal exposure model.

For PM_{10} , only one study on railway transport exists by Praml & Schierl (2000) undertaken for tram lines in Munich. The study monitored concentrations for five different lines between $71\mu\text{g}/\text{m}^3$ and $279\mu\text{g}/\text{m}^3$ with an average of $183.41\mu\text{g}/\text{m}^3$. Unlike most other rail transport, tram lines often run along roads and may as such be influenced by road sources. An estimate for exposure in railway transport based on concentrations inside trams may not, therefore, represent concentrations in other railway transport systems well. The tram lines studied by Praml & Schierl (2000) were therefore further investigated. A detailed description of each line, provided online, reveals that only parts of the five investigated tram lines run in direct vicinity to road traffic (http://de.wikipedia.org/wiki/Stra%C3%9Fenbahn_M%C3%BCnchen, accessed: 21st May 2014).

Other parts of the lines are separated from direct influences of traffic sources (sometimes by using completely separated routes, sometimes running in parallel to roads, separated by sidewalks and vegetation). The average monitored concentration of $183.41\mu\text{g}/\text{m}^3$ was consequently considered to be representative of concentrations in rail transport and was as such applied to estimate PM_{10} concentrations for times in railway transport.

The estimates of $20.64\mu\text{g}/\text{m}^3$ for $\text{PM}_{2.5}$ and of 183.41 for PM_{10} selected for exposure estimates in railway transport differ substantially from each other. To some extent this may reflect a particularly high difference between size fractions for the railway transport ME. Vijayan & Kumar (2010) have for example shown for in-bus exposure that resuspension caused by passenger movement raises coarse particle concentrations in particular. It is however also possible that the estimate for PM_{10} is influenced to some extent by road traffic sources near the tram lines. This may have increased concentrations.

5.2.7. Discussion

In this section a method for monitoring along routes using the ESCAPE-LUR model was explained and adjustments from ambient concentrations to exposure in traffic MEs, as well as estimates for exposure in railway transport have been introduced.

Several limitations apply to the adjustment of in-traffic MEs (cycling, bus, and car) by ratio:

- Adjusting in-traffic exposure for horizontal displacement from the sidewalk, does not adjust for vertical displacement. Concentrations underlying the ESCAPE-LUR model have on several occasions been recorded above breezing height. It is therefore possible that concentrations at the sidewalk are under-predicted by the model.
- The adjustment ratios only correct for the total difference between roadside and traffic mode concentrations, they do not account for added concentration variability closer to traffic sources or for variability caused by in-vehicle sources.
- It has been shown that the adjustment ratios for cycling and in-bus exposure vary temporally, as well as spatially. These differences are not accounted for in the application of a mean ratio.
- The adjustments calculated for this thesis from monitoring data were based on relatively small datasets. Bigger datasets may have provided ratios that better represent exposure in these microenvironments.

For the in-railway estimates further clear limitations exist.

- The fixed estimates do not allow for any variability during times in railway transport.
- For the PM_{2.5} estimate, a mean concentration for times in railway transport above ground had to be calculated from provided concentrations for the whole journey (above and below ground). For this calculation an assumption had to be made that 50% of the subway line was above and 50% below ground.
- The estimates are not representative of in-tunnel exposure
- The estimate from the literature used for PM₁₀ is based on concentrations in tram lines. The tram lines used for this study are close to traffic for parts of the route, which may have increased concentrations inside the tram. An estimate based on in-tram exposure may not be representative for exposure in other railway transport.

Both the adjustment ratios, as well as the estimates for in-railway transport introduced above are only proxies for the exposures in the different transport MEs, which cannot reflect the full concentration variability in these MEs. Their application was nonetheless considered crucial to avoid underprediction of in-transport exposure and in consequence exposure misclassification.

Application of in-transport adjustments has been undertaken for other personal exposure models. Several studies have used an in-car adjustment to adjust for all in-vehicle MEs, including buses (Borrego et al. 2006; Gulliver & Briggs 2005; Briggs et al. 2005). It has however been shown in chapter 2, that the two microenvironments differ substantially, as parameters, such as infiltration and indoor sources are different. One study, by Gerharz et al. (2009) used a roadside to in-bus ratio by Praml & Schierl (2000) as adjustment. The in-bus to roadside ratio by Praml & Schierl (2000) is however based on a nearby roadside monitoring station instead of simultaneously monitored concentrations along the same route. For the development of a personal exposure model in this thesis, the development of an in-bus adjustment based on simultaneously monitored concentrations along the same route was considered to better reflect the in-bus to roadside difference.

Only one other study by Gariazzo et al. (2014) included railway transport into a personal exposure model. The study applied an indoor-outdoor ratio developed for home locations to model PM_{2.5} exposure in trains and metros. Home indoor environments however substantially differ from in-railway environments in several ways: 1) Surrounding ambient concentrations differ (due to the breakware etc. around trains), 2) the indoor sources differ (usually more people, as well as frequent movement of passengers inside trains), and 3) the permeability of the walls differ (usually frequent door openings in local trains). This approach was therefore not considered an improvement to the estimates used to predict in-railway exposure for this thesis.

The selected in-transport adjustments and estimates are combined with other components of the personal exposure model in chapter 6.

5.3. Modelling Home Indoor Concentrations

5.3.1. Introduction

Most people spend the majority of their time indoors (Schneider et al. 2004). Good estimates for indoor concentrations are therefore important for personal exposure modelling. Indoor concentrations during personal exposure are commonly adjusted using indoor models from the literature. Up to this point very few attempts have been made to evaluate indoor model performance or to compare existing models. In this chapter three different types of indoor models have been applied to predict indoor concentrations for Home A (see chapter 3). The model predictions and performances were evaluated and results were compared between models.

The models compared here are A) indoor-outdoor (I/O) ratio, B) regression models in three variations, and C) mass-balance (MB) models in four variations. All three types of models have been described in detail in chapter 2. An indoor-outdoor ratio has been applied as it is one of the simplest and most commonly applied indoor models used in exposure modelling. A study by Hodas et al. (2014) for $PM_{2.5}$ has shown that I/O ratios do not reflect concentration variability in homes well and that more complex models should be applied to avoid exposure misclassification. Regression models represent an easily applicable, yet largely underdeveloped type of indoor model. As described in chapter 2, one of the first regression models based on indoor situations (sleeping, cooking etc.) was developed by Jamieson (2012). A set of three regression models developed by Jamieson (2012) was applied, the first version only includes outdoor concentration and occupancy, the second distinguishes between types of source activity during occupancy, and the third combines a time model (for hours of days and days of weeks) with the second model. The regression models were included in this comparison between indoor models as they might provide an alternative to the other, more commonly applied models. The third type of model applied here was the MB model. MB models are based on physical parameters that influence input and output to and from indoor concentrations. As described in chapter 2, many studies have developed and validated model parameters for MB models. Little evidence is however available about the performance of these models and how they compare to other approaches. Jamieson (2012) suggests in her work that the physical principles underlying the MB model reflect indoor concentrations better compared to regression models. Several MB models of different complexity were applied to evaluate and

compare the approach. The most simplistic version applied here only uses outdoor concentrations to model indoor particles. This MB model may provide an alternative to the commonly applied I/O ratio. The second MB model version includes source emissions for cooking events from the literature. This version is one of the most commonly applied MB models (Hoek, Kos, et al. 2008; Hänninen et al. 2004; Ferro et al. 2004; Nazaroff 2004).

As a further development of the MB model, a resuspension parameter for “active occupancy” (people at home, not sleeping) has been calculated and was applied in a third model version. A parameter for “hour after cooking” has furthermore been developed and has been added to the third model. These four versions of MB models were consequently applied and results were described, examined and compared together with results from the I/O ratio and regression models.

A particular focus in the interpretation of results was the investigation of peak concentrations and how well different models were able to reflect these. As described in chapters 2 and 3, a typical feature of home indoor environments is high source (including resuspension) related concentrations that typically last from a few minutes up to a few hours. Short-term peak concentrations may cause adverse health effects, as well as influence the mean concentrations for longer time periods. In homes without ETS (environmental tobacco smoke), the main causes for these peaks are cooking and to some extent resuspension caused by people’s movements. Simple models such as the indoor-outdoor ratio or MB-infiltration models do not distinguish between indoor source activities and may therefore not reflect indoor concentrations well, as suggested by several studies (see chapter 2). The popularity of these approaches rather lies in their ease of application and low data requirements. These models are applied here as a comparison between each other and as a reference to the more detailed models. MB and regression models can include parameters on source activities, but it is not known how well they actually predict source related concentrations. Results from this model comparison provide important information about indoor models, which is crucial to the understanding of predicting indoor concentrations in the context of personal exposure model development.

Datasets and their application:

The dataset collected for home A is used for direct comparison of the models, as described in chapter 3. The dataset includes hourly monitoring concentration of $PM_{2.5}$ and PM_{10} (averaged from minute concentrations) for home indoors and home outdoors for 39 days (collected between April 2010 and February 2011). In addition to the monitoring data, the parameters of the regression and the MB model require diary data at a minute resolution, with information on people’s movements

and cooking times. Diary data has been collected for measurements of home indoor concentrations, as described in chapter 3. An additional dataset was used to calculate a parameter for the MB model using concentrations from indoor and outdoor monitoring for a subset of data collected at homes B, C, and D. The dataset is described below in further detail. The regression models applied here required log-normal transformation of outdoor concentrations, as the regression coefficients had been calculated with log-normal transformed data (Jamieson 2012). The outdoor concentrations were therefore log-transformed for the calculation of the model. The transformation was reversed for resulting predictions of indoor concentrations before estimates were compared with indoor monitored concentrations.

5.3.2. Indoor-Outdoor Ratio

Indoor-outdoor ratios (I/O ratios) have been calculated from the difference between indoor and outdoor concentrations by many studies and are commonly applied in personal exposure studies (Dimitroulopoulou et al. 2001; Borrego et al. 2009; Jensen 1999; Hänninen, Karppinen, et al. 2005), as described in chapter 2. An I/O ratio was applied from the literature in order to evaluate and compare the results from this simplistic, but often applied form of indoor model with more complex models. For this application I/O ratio results from large datasets (with multiple houses) were preferred. Few large studies which calculated I/O ratios exist (see chapter 2). For $PM_{2.5}$ a ratio from the EXPOLIS study for Helsinki (Koistinen et al. 2001) was chosen. It is one of the largest studies, including 135 homes of non-smokers. Additionally it is based in Europe with a housing stock similar to British housing (compared to other big USA-based studies). For PM_{10} , no large European study exists; therefore results were taken from the USA-based PTEAM study (Özkaynak et al. 1996). The PTEAM study calculated indoor-outdoor ratio from 735 homes. The applied indoor-outdoor ratios are:

- For $PM_{2.5}$: 0.86
- For PM_{10} : 1

The above factors have been multiplied with the outdoor concentrations monitored at Home A in order to model indoor PM for comparison with other models.

5.3.3. Regression Models

Using a regression equation to model influences on indoor air has not been investigated by many studies (see chapter 2). The regression approach generally starts with the outdoor particle

concentration as a first variable, but has the capacity to then include further variables for different situations and sources influencing the indoor air (occupancy, cooking events, sleeping/waking times etc.). A model by Jamieson (2012) was chosen for application for several reasons:

- A) Several regression models were available from Jamieson (2012), which reflect different levels of model complexity. This allows the investigation between different variations of this approach.
- B) The models were developed for the same baseline time period (hourly) used in this study.
- C) The model has been developed for a flat in London providing a similar environment to home locations modelled by this thesis.
- D) All data requirements including detailed diary information for the model by Jamieson (2012) could be met.

Some limitations apply, namely that the monitoring dataset for model development only included data from one home for 26 days (819 hours); and only limited validation was undertaken (1 home, 9 days – 100 hours). The monitoring for model development has been undertaken by two GRIMM dust monitors, model 1.105 (GRIMM Aerosol Technik GmbH & Co., Ainring, Germany). Uncalibrated concentrations collected by these monitors have shown to not be fully reference equivalent (Walden et al. 2010), which may have altered results for the regression equation. Additionally, some indoor influences, such as window openings and concentration increase after cooking were not included in the regression equations. Yet these models provide the furthest developed regression model set available for application (see chapter 2)

The regression model by Jamieson (2012)

The regression models by Jamieson (2012) is in this chapter also referred to as ‘the regression models’, or abbreviated to REG-J. Concentrations for model development have been collected at one residence between September 2004 and January 2005, data for validation was simultaneously collected for indoor and outdoor locations during August 2007 at a different location. The model has been calculated for fine PM (PM_{1-3.5}) and for PM₁₀. The range of fine PM differs with particle sizes from 1µm to 3.5µm from the fine PM particle sizes used for this thesis (up to 2.5µm). An application of the fine PM model was still considered possible as the size fractions overlap considerable (between 1µm and 2.5µm). It is however possible that slight differences in physical properties, such as differences in particle deposition, may affect model fit. The model was developed using linear regression. Explanatory variables were entered in an automated stepwise procedure, inclusion (and

removal) of variables was restricted by an entry at $p < 0.05$ and a removal at $p > 0.10$ (p =significance). Several model phases were undertaken in the study:

Model 1 models ambient concentrations to reflect particles at the home façade.

Model 2 is a regression model including outdoor PM and dichotomous variables indicating indoor occupancy and activities. For **model 2a** only one variable on occupancy (at home or not at home) was added in the model. For **model 2b** more detailed variables were added for sleeping, sitting, moving, and cooking. Hourly dichotomous variables for the model were calculated according to a specific hierarchy: cooking > moving > sitting > sleeping.

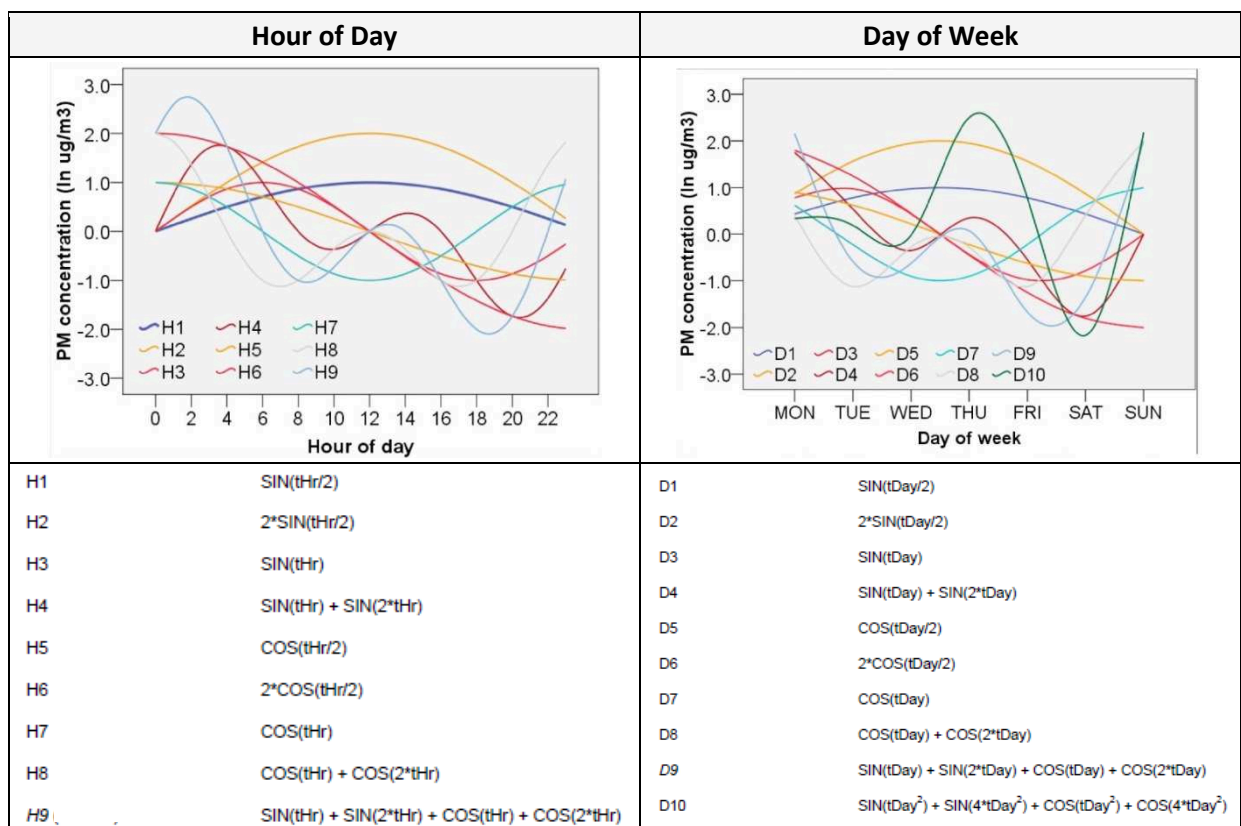


Figure 40: Trigonometric time cycles for hours of the day and days of the week. Source: Jamieson (2012)

TEMP Model: in preparation for model 3b a time model was produced in a regression against the residuals of model 2b. To calculate time variables, first an hour of day and day of week time cycle was calculated, as follows:

Equation 7: Position of hour in a 24-hour cycle (after: Jamieson (2012)):

$$t\text{Hr} = (2\pi * \text{Hour})/24$$

Equation 8: Position of individual day in a 7-day time cycle (after: Jamieson (2012))

$$tDay = (2\pi * Day)/24$$

with tHr the position of individual hour in a 24-hour time cycle, $tDay$ the position of each individual day in a 7-day time cycle, π the equivalent to 3.14159265, $Hour$ the hour of day (0-23), and Day the day of week. The time cycles were then calculated into a range of sinus and cosinus functions to reflect time patterns across days and weeks, as presented in Figure 40. These trigonometric functions were introduced as potential input variables into a stepwise regression. The time variables that entered the regression were: H3, H4, H8, and D8 for the $PM_{2.5}$ model, and H1, H3, H5, H7, and D8 for the PM_{10} model.

Model 3b: For model 3b input variables for the regression consisted of results modelled by model 2b, as well as results modelled by the time model.

The resulting model equations in Jamieson (2012) for models 2a, 2b, TEMP, and 3 for fine PM and PM_{10} are presented in Table 47, including R^2 adjusted results and SEE (Standard Error of the Estimate).

		Equation	R^2 adj	SEE ¹
Fine PM	2a	$-0.564 + \text{outdoor} * 1.158 + \text{occupancy} * 0.620$	0.233	1.83
	2b	$0.207 + \text{outdoor} * 0.803 + \text{sitting} * 0.745 + \text{moving} * 0.925 + \text{cooking} * 1.313$	0.375	1.72
	TEMP	$-0.005 + H3 * 0.132 + H4 * -0.139 + H8 * 0.119 + D8 * 0.064$	0.097	0.52
	3b	$0.196 + \text{model2b} * 0.908 + \text{TEMP} * 1.048$	0.445	1.67
PM_{10}	2a	$-2.142 + 1.371 * \text{outdoor} + 1.212 * \text{occupancy}$	0.418	2.02
	2b	$-0.399 + 1.003 * \text{outdoor} + \text{sitting} * 1.366 + \text{moving} * 1.625 + \text{cooking} * 2.019 + \text{sleeping} * 0.548$	0.530	1.88
	TEMP	$1.794 + (-2.820 * H1) + (-1.122 * H7) + (-0.498 * H5) + (0.420 * H3) + (0.075 * D8)$	0.152	1.78
	3b	$0.391 + 0.880 * \text{model_2b} + 1.095 * \text{TEMP}$	0.615	1.77

Table 47: Indoor regression model equations for fine PM and PM_{10} , as by Jamieson (2012); R^2 adjusted and SEE (Standard Error of the Estimate) are added, ¹ un-log applied.

Validation was undertaken with data collected at a different home (with the same occupants) for 100 hours for models 2b and 3b. R^2 Results for the application of the validation dataset were however low for the fine PM fraction with 0.055 for model 2b and 0.022 for model 3b. For PM_{10} , results for the validation were better with an R^2 of 0.559 for model 2b and an R^2 of 0.460 for model 3b.

The models presented above show an interesting approach to include source related activities into modelling indoor particles. For the fine PM model the difference in particle size ($PM_{0.5-3.5}$, not $PM_{2.5}$) and the low results for model validation however raise some concerns for model

transferability. The PM_{10} models performed generally better in model development as well as for validation.

Model application

For the application in this thesis outdoor concentrations monitored at the house façade were directly entered into models 2a and 2b. Variables on diary data could be precisely reproduced, as all relevant information had been collected during monitoring campaigns. Models 2a, 2b, and 3b were calculated as described, results are presented below in comparison with the other model types.

5.3.4. Mass-Balance Models

A detailed description of MB models and parameters has been presented in chapter 2. In the following section four MB models including no source activity (MB1), as well as one, two and three parameters for source activities (MB2, MB3a, MB3b) are applied to model indoor concentrations. In addition to parameters from the literature, two source parameters are calculated and applied.

First, an infiltration model, the most simplistic form of an MB model was applied. An infiltration model by Hänninen et al. (2004), as described in chapter 2, Equation 2, was selected for this application. The infiltration model is in following paragraphs referred to as MB1.

Other MB models applied in this thesis include source activities as an additional parameter. One of the most widely recognised MB model version of this type by was chosen for application, based on Koutrakis et al. (1992) and as applied by Gerhartz et al. (2009), and introduced in chapter 2, Equation 3. Source including MB models do not specify the type of sources, potential source parameters are therefore discussed below. All four MB model versions are applied and compared for predictions of indoor concentrations at home A.

Parameter Selection of Penetration (p), Deposition (D), and Air Exchange (α)

In order to apply the MB models, estimates of parameters, as introduced in chapter 2, were selected from the literature. Parameters calculated for MB models are summarised in Table 48. The three parameters used in infiltration models: penetration efficiency (p), deposition (D), and air exchange rate (α) form the base for the calculation of all MB models. These three parameters have been calculated by a few large studies whose estimates have since been applied by others. For both $PM_{2.5}$ and PM_{10} results from the PTEAM study were used here for penetration efficiency (p), as well as for

deposition (D) rate. The PTEAM study includes 735 residences, making it by far the biggest study providing parameter estimates. Its results have been applied in MB models by several other studies before (Gerharz et al. 2009; Hänninen et al. 2004; Wallace et al. 1996). The study did however include residences with environmental tobacco smoke (ETS), which does not match the study home for this application. In addition, the PTEAM study is based in the USA. As mentioned before, housing stock in the USA differs from British homes, which may have influenced the calculated parameters. For the air exchange rate results from the BRE (Building Research Establishment) study were used, a large British study including 385 homes were therefore chosen, instead of further results from the US-based PTEAM study. The BRE study focuses on air tightness of homes, its air exchange estimate has been applied in an MB equation before by the INDAIR study (Dimitroulopoulou et al. 2006). The selected main parameter estimates for PM_{2.5} are as such: $p=1$, $\alpha = 0.7$, and $D = 0.39$. Parameter estimates for PM₁₀ are: $p=1$, $\alpha = 0.7$, and $D = 0.65$.

	Study	n	p	α	D	Ei Cook	Limitations
PM _{2.5}	Allen et al. 2003	44	0.94	0.54	0.20		USA based
	BRE study (Perera & Parkins 1992)	385		0.7			
	Fogh et al. 1997	2			1.73		
	EXPOLIS study Hänninen et al. 2004	158		0.7 – 1.3 (4 cities)			
	He et al. 2004	15				0.11, (0.84 ¹)	Australia based
	Koutrakis et al. 1992	394	0.84	0.51			Winter only, ETS
	Long et al. 2001	9	1.11 (S), 0.54 (W)		0.15 (S), 0.1 (W)		USA based
	Olsen & Burke 2006	37			0.27	36 ²	
	PTEAM 1 & 2 (Özkaynak et al. 1996 & Özkaynak et al. 1997)	735	1	1.01 (n=135)	0.39		ETS included, USA based
	PTEAM 3 (Wallace et al. 1996)	178	1			1.7	ETS included, USA based
PM ₁₀	Abt et al. 2000	4	0.12-0.53				USA based, PM _{0.7-10}
	BRE study (Perera & Parkins 1992)	385		0.7			
	PTEAM 1 (Özkaynak et al. 1996)	135	1	1.01	0.65		ETS included; USA based
	PTEAM 2 (Özkaynak et al. 1997)	175		0.97			ETS included; USA based
	PTEAM 3 (Wallace et al. 1996)	178				4.1	ETS included; USA based

Table 48: Summary of key studies, which calculated parameters for mass-balance models. n = number of residence; p = penetration efficiency; α = air exchange rate per hour; D = deposition in mg/m³ per hour; k = decay in mg/m³ per hour; Ei Cook = Cooking emission rate in mg/m³ per min; S = summer, W = winter, ETS = Environmental Tobacco Smoke ¹cooking test with onions for specific ventilation settings. Parameters applied in this thesis are highlighted in red.

Parameter selection of cooking source strength

Compared to the main parameters (p , D , and α), described above, estimates on indoor sources (and resuspension) parameters are less well established. Source emission estimates, called E (or E_i) are

mainly available for ETS and for cooking (E_{cook}). A parameter for ETS can however not be validated in this application, as home A was a non-smoking home. ETS parameters are therefore not further investigated in this context. A study by He et al. (2004) calculates two different estimates for cooking source strength, one as part of a personal monitoring campaign including 15 homes, the other as part of small experimental cooking campaign (cooking onions under specific ventilation conditions). The study uses short-term concentration results to calculate the parameter from which cooking source estimates could be derived directly. Periods of cooking in this study were considered similar to those of the personal monitoring campaign in the study by He et al. (2004). The cooking source strength estimate for $\text{PM}_{2.5}$ by He et al. (2004) of $0.11\text{mg}/\text{min}$ was therefore applied in this thesis.

The much larger PTEAM study also provides cooking source strength parameters, but collected concentration in passive samplers for 12 hour time periods. Source strengths could then only be calculated indirectly, by assuming estimates of all other influences on indoor concentrations for the period of each sample time (12h). One other study by Olson & Burke (2006) provides results for cooking source strength for a dataset of 37 homes. The mean source strength given is however unrealistically high with $36\text{mg}/\text{min}$ (this would frequently produce concentrations during cooking events of several $10,000\mu\text{g}/\text{m}^3$). The estimate was therefore not considered for further application, as it may include a calculation error.

For PM_{10} , the only transferable estimate for cooking source strength is provided by the PTEAM study (Wallace 1996), with an estimate of $4.1\text{mg}/\text{m}^3/\text{min}$. This estimate was therefore selected for application in this thesis. Other studies which also applied this parameter estimate are Dimitroulopoulou et al. (2006) and Zidek et al. (2003). As mentioned above, the PTEAM study provides a large dataset (178 residences for cooking source estimates). Several limitations however apply, namely the calculation of source strength from 12h average concentrations, the location of the study in the USA, and the inclusion of ETS. One other estimate for PM_{10} cooking source strength was calculated by Abt et al. (2000). Several limitations however apply to that study. 1) Concentrations for $\text{PM}_{0.7-10}$ were recorded instead of PM_{10} . 2) Concentrations were inconsistently recorded for 5 and 15 minutes. 3) Results are given for volume of $\mu\text{m}^3/\text{cm}^3$ (which can only be converted to roughly approximate $\mu\text{g}/\text{m}^3$, see Olson & Burke (2006) and Wallace et al. (2006). 4) The study was small including only four homes. For these reasons the results by Abt et al. (2000) were not applied here as parameters. Parameter estimates of $0.11\text{mg}/\text{m}^3$ for $\text{PM}_{2.5}$ and $4.1\text{mg}/\text{m}^3$ for PM_{10} were subsequently transformed into μg and entered into Equation 3, as source emission parameters, which can be written as follows:

Equation 9: MB model 2

$$C_{in} = \frac{C_{out} * p * \alpha + \frac{E_{cook} * cookmin}{V}}{\alpha + D}$$

Where, C_{in} = indoor concentration, C_{out} = outdoor concentrations, α = air exchange rate, D = particle deposition, p = the penetration factor, V = each resident's volume in m^3 , and t = time of source emission. E_{cook} is the cooking source emission strength and $cookmin$ the minutes of cooking per hour. In order to improve the currently available MB model two additions were calculated by this thesis: 1) a source strength for active occupancy, 2) a parameter adjusting for the effect of particles during the hour after cooking.

Calculating a Parameter for Active Occupancy

A main cause of re-suspension of particles indoors is movement of occupants (see chapter 2). The particles added into the indoor air through resuspension effectively act as an indoor source and is therefore and referred to here as an emission source. To the author's knowledge, no other study provides transferable estimates for active occupancy or people's movement that could be included as source strength in the MB models (results provided by Abt et al. (2000) were not considered suitable, see above). It was therefore decided to calculate a parameter for active occupancy using the MB equation.

Data preparation:

Indoor-outdoor concentrations monitored at homes B, C, and D were used to calculate the increment in PM concentrations associated with different levels of occupancy indoors. Indoor and outdoor data for the three homes have been pooled to reflect different homes and differences in people's behaviour in the data. The dataset consists of 19 days (home B: 8 days, home C: 7 days, home D: 4 days), monitored between June and September 2011. Source activity can be calculated with the MB equation if all other parameters are known. The dataset was therefore cleaned from any other main source activity in order to reduce the number of additional parameters in the equation. The only other major source was cooking. Hours with cooking activity were removed from the dataset, as well as a potential cooking lag time (see chapter 2 and 3) of three hours after cooking. One exception applied: A cooking event of one minute for which only the actual hour of cooking was removed.

Occupancy and activity patterns were simplified as someone on the main floor and active (not sleeping) and were recorded as number of minutes per hour. Only having one activity category provides the option to use this parameter for homes without recorded time-activities in future applications, by making assumptions about when people are at home and active. These assumptions can for example be based on widely available data, such as employment rates. After data preparation altogether 51 hours with people activity were analysed for the parameter.

Method:

From the MB model (Equation 3) the following equation can be derived for source emission related indoor concentration (one source type only) by simple mathematical transformation, following an approach by Ferro et al. (2004):

Equation 10: Emission source strength.

$$E = \frac{((C_{in} * (\alpha + D)) - (C_{out} * p * \alpha)) * V}{t}$$

With E being the source emission, and as before: C_{in} = indoor concentration, C_{out} = outdoor concentrations, α = air exchange rate, D = particle deposition, p = the penetration factor, V = each resident's volume in m^3 , and t = time of source emission. Specified for active occupancy or people's movement (E_{move}) the equation can be written as follows:

Equation 11: Active occupancy as source activity.

$$E_{move} = \frac{((C_{in} * (\alpha + D)) - (C_{out} * p * \alpha)) * V}{movemin}$$

With E_{move} as source emission equivalent of people's active occupancy and $movemin$ as the time people spent at home not sleeping in minutes per hour. The equation was subsequently calculated with C_{in} and C_{out} from monitored hourly indoor and outdoor concentrations, V as the volume of each respective home, and all other parameters as selected from the literature, see above (Table 48). The source strength concentration was then calculated (per cubic metre per minute), and results were transformed into mg (as is the convention for source strength).

Results:

The source strengths were calculated for PM_{2.5} and PM₁₀ and results are presented in Table 49. Mean source strength is 0.0529mg/min for PM_{2.5} and 0.350mg/min for PM₁₀ (both calculated for 1 m³). The SDs are high in comparison to the mean, with 0.176 and 1.304 for PM_{2.5} and PM₁₀ respectively.

	Mean	Standard Deviation	Range
PM _{2.5}	0.0529	0.176	0.562
PM ₁₀	0.350	1.304	3.913

Table 49: Basic statistics for source strength equivalent of active occupancy indoors for PM_{2.5} and PM₁₀.

Substantial variability around the mean was however expected, as the parameter calculated for people activity includes different forms of activities, such as sitting, walking, tidying, or children playing. The mean results are used as an estimate of average concentration increase for times people are at home and not sleeping. As such they were included in the MB model and the model version with included active occupancy (as based on Equation 3) was called MB3a and can be written as follows:

Equation 12: MB model 3a

$$C_{in} = \frac{C_{out} * p * \alpha + \left(\frac{(E_{cook} * cookmin)}{V} + \frac{(E_{move} * movemin)}{V} \right)}{\alpha + D}$$

With parameters as described for Equation 11, and E_{move} as source emission equivalent of active occupancy and $movemin$ as the time people are at home and not sleeping in minutes per hour.

Calculation of 'hour after cooking'

Every hour modelled by a MB model starts in theory with zero pollution to which pollution is added (from modified outdoor pollution and from source activities) and removed (decay and out-filtration). This ignores the fact that pollution levels are maintained over time and becomes particularly problematic after high intensity source activities, such as cooking. It has been suggested by Schneider et al. (2004) to add the concentration from the previous hour to each modelled hour. An attempt to include cooking lag times into an indoor model has been made by Abt et al. (2000), who included a cooking lag time (minutes cooked in the previous 20min time slot) in a regression for source activities, calculated for PM sizes up to PM_{0.5}.

The slow decay of concentrations after cooking events has been recognised in the literature (see chapter 2) and shown for data from this thesis in chapter 3. The second addition to the MB model was therefore made by calculating a parameter as part of this thesis for the hour after cooking activities by using the MB model. While cooking decay times vary, the effect on the first hour after cooking is generally most pronounced and most recognised in the literature (Olson & Burke 2006; He et al. 2004; Abt, Catalano, et al. 2000). A focus on the first hour after cooking was therefore undertaken.

Calculation:

A simple calculation was undertaken by this thesis to add cooking related concentrations from the previous hour. For the hour after cooking (called cooklag) the cooking source related concentrations from the previous hour were added, reduced by the hourly particle loss, which can be calculated from parameters in the MB model (air exchange and deposition). To start with, the concentrations added by cooking source activity (using parameters described above) during the actual cooking hours were calculated. To calculate only cooking related indoor concentrations the part of the MB equation (Equation 3) related to source concentrations was extracted from Equation 9 (following calculations by Hänninen et al. (2004)), which then reads as follows :

Equation 13: Indoor concentrations related to cooking emissions

$$C_{\text{cook}} = \frac{\frac{E_{\text{cook}} * \text{cookmin}}{V}}{\alpha + D}$$

As such, the indoor concentrations related to cooking emissions (C_{cook}) are calculated from the cooking source strength (E_{cook}) multiplied by the minutes of source activity (cookmin). These are divided by the home volume (as source emission strength is given for 1m^3). The loss of indoor concentrations (per hour) is calculated in the denominator from particle deposition and air exchange. For the concentrations added to the indoor environment during the next hour (C_{cooklag_h}) this means that the concentrations related to cooking in the previous hour ($h-1$) can simply be added as another input to the indoor system reduced by the hourly loss, see Equation 14:

Equation 14: Indoor concentration related to cooking in the previous hour

$$C_{\text{cooklag}_h} = \frac{C_{\text{cook}_{h-1}}}{\alpha + D} \Rightarrow C_{\text{cooklag}_h} = \frac{\left(\frac{E_{\text{cook}_{h-1}} * \text{cookmin}_{h-1}}{V} \right)}{\alpha + D}$$

For the calculation of the MB3b model, the cooking concentrations for the previous hour have been added similar to an additional source. The added concentrations are already scaled by minutes of cooking and house volume. The final model, called MB model 3b reads as follows:

Equation 15: MB model 3b

$$C_{in} = \frac{C_{out} * p * \alpha + \left(\frac{(E_{cook} * cookmin)}{V} + \frac{(E_{move} * movemin)}{V} + C_{cook_{h-1}} \right)}{\alpha + D}$$

Parameters of Equation 15 have been described above. All final parameter estimates, as used in the models MB1, MB2, MB3a, and MB3b have been summarised in Table 50 for further reference. All four models described above were subsequently applied to estimate indoor concentrations for Home A.

	C _{out}	p	α	V	k (D)	cook (mg/min/m ³)	cooklag (mg/h/m ³)	move (mg/min/m ³)
PM _{2.5}	monitored	1	0.7	per home	0.39	0.11	Calculated from cook _{h-1}	0.0529
PM ₁₀	monitored	1	0.7	per home	0.65	4.1	Calculated from cook _{h-1}	0.350

Table 50: Summary of all parameter estimates applied in mass-balance models.

5.3.5. Results for the Prediction of Particle Concentrations in Home A

All indoor models described above have been applied to calculate hourly predictions for 39 days monitored at home A. In the following paragraphs predictions for indoor concentrations for the home are compared. Basic statistics of both the monitored indoor concentrations and the modelled concentrations are summarised in Appendix I, including bar charts for mean concentrations by source related activities for modelled and monitored concentrations. Scatter plots of all models against the monitored concentrations are shown in Figure 41 for PM_{2.5} and in Figure 42 for PM₁₀. In the scatter plots source related categories are presented in different colours (MB and regression models only). All plots are presented using log scales for both axes (to accommodate for high concentrations for example during cooking times). Results for model performance parameters (as introduced in chapter 3) are presented in Table 51 for PM_{2.5} and in Table 52 for PM₁₀.

Results for PM_{2.5}

The results show that general performance of all models is poor with very low R² ranging from 0.005 (I/O Ratio and MB1) to 0.051 (Model MB3b). The results for the Index of Agreement (IA) range from 0.083 (I/O ratio) to 0.251, showing weak model fit (good model fit usually considered for IA > 0.5).

The results for the Normalised Mean Square Error (NMSE) are very high ranging between 8.429 and 17.269. The Fractional Bias shows underprediction for all but the REG-J 2a model. FB results for this analysis lie between -0.600 and 0.103. All models as such under- and overpredict by less than a factor of 2 (FB results under -0.67 or over 0.67 are equivalent to under- or overpredictions by a factor of two, see chapter 2). The scatter plots reveal that at a log10 scale, monitored and modelled concentrations for the majority of data points show a linear relationship, which is not reflected in the weak results of the model performance parameters. This relationship is however only evident for lower concentrations.

Results for the indoor-outdoor ratio show the lowest performance parameters of all predictions with a low R^2 of 0.005, a low IA of 0.083, a very high NMSE of 12.757 and general underprediction of the data indicated by an FB of -0.324.

For the regression models results for model performance parameters are also generally low, but show a very small improvement in model performance from model 2a to 2b with an improvement for R^2 (from REG-J 2a: 0.007 to REG-J 2b: 0.035) and for IA (from REG-J 2a: 0.120 to REG-J 2b: 0.147). The difference between the models is likely to better reflect indoor sources. The error is however higher for REG-J model 2b with an NMSE of 10.173 compared to 8.429 for REG-J model 2a. The REG-J model 2a overpredicts with an FB of 0.103, the REG-J model 2b underpredicts with a higher misprediction of -0.144. The third REG-J model 3b shows almost similar results to the REG-J model 2b with a slightly higher NMSE of 10.434 and slightly more underprediction with an FB of -0.169. The I/O ratio as a third model type shows one of the lowest performances with an R^2 of 0.005, an IA of 0.083, an NMSE of 12.757 and an FB of -0.324. The scatter plots for the regression model show some difference between models 2a and 2b. JEG-J2a shows no differentiation between indoor situations, for model 2b. Some differences, for example between cooking and sleeping, are visible.

Model	R^2	IA	NMSE	FB
I/O Ratio	0.005 *	0.083	12.757	-0.324
REG-J 2a	0.007 **	0.120	8.429	0.103
REG-J 2b	0.035 **	0.147	10.173	-0.144
REG-J 3b	0.036 **	0.147	10.434	-0.169
MB1	0.005 *	0.089	17.269	-0.600
MB2	0.018 **	0.110	16.245	-0.559
MB3a	0.028 **	0.194	9.634	-0.067
MB3b	0.051 **	0.251	9.107	-0.044

Table 51: Model performance parameters (R^2 , IA, NMSE, and FB) for predictions of indoor-outdoor (I/O) ratio, regression models (REG-J) and mass-balance models (MB) for $PM_{2.5}$ concentrations at Home A. * $p < 0.05$; ** $p < 0.01$

Model performance parameters of the MB models show better results with increased number of included source parameters. For the MB models one parameter was added for each subsequent model: MB1, to MB2, to MB3a, to MB3b (all other parameters stayed the same). The performance difference between models can therefore directly be connected to the added parameters. Of the four MB models, the lowest performance (R^2 of 0.005, an IA of 0.089) and highest error (NMSE of 17.269) was calculated for MB1 with no indoor source emissions included. For model MB2 the added cooking source strength very slightly increased performance (R^2 of 0.018 and IA of 0.110). The new parameter of “active occupancy” (called people movement in the graph) was added as a source in MB model 3a and improved predictions further (R^2 of 0.028 and IA of 0.194). A more pronounced change can be seen for the NMSE, which is for model MB3a much lower with 9.634 compared to model MB2 with 16.245. In addition, underprediction improved from an FB of -0.559 for MB2 to an FB of -0.0668 for MB3a. A further improvement of model performance can be seen after including “hour after cooking” for MB3b. The R^2 shows in comparison to the other MB models an increase to 0.051, the IA is improved to 0.251. The NMSE and FB are both very slightly improved to an NMSE of 9.107 and an FB of -0.0440. For the scatter plots the addition of source emission parameters for different MB models can be followed throughout the scatter plot sequence from left to right. Each of the three added parameters (cooking, active occupancy, and hour after cooking) increased the relevant concentrations. The scatter plots reveal the actual variability within these source related situations, which is not reflected by the deterministic parameters of the model.

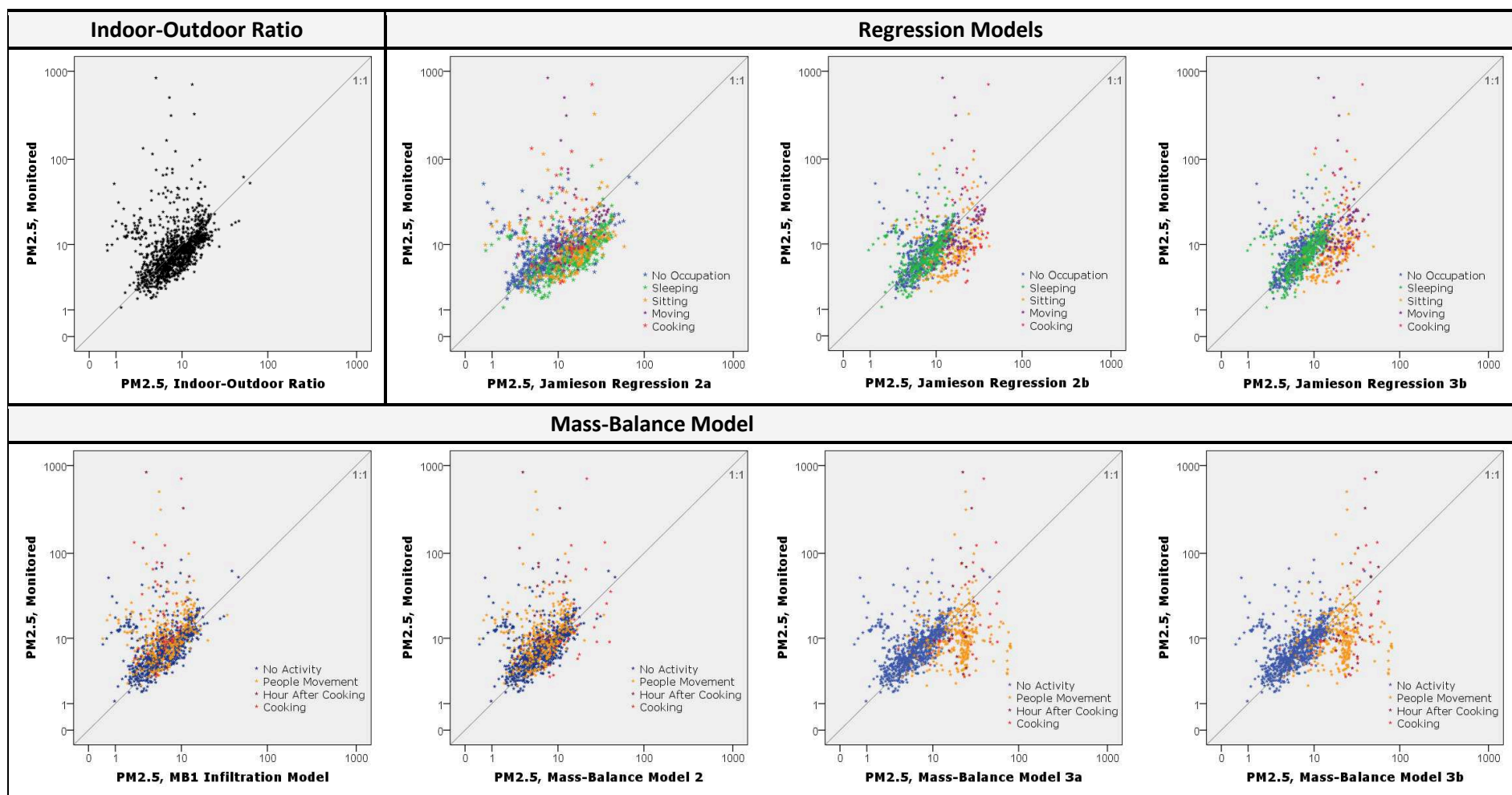


Figure 41: Modelled against monitored indoor concentrations for PM_{2.5} (in µg/m³) using indoor-outdoor ratio, regression models (2a, 2b, 3), and mass-balance models (1, 2, 3a, and 3b). Different source activities are added. All presented plots use log scales both for X and Y axes. 1:1 reference line is added to each graph.

Results for PM₁₀

Results for model performance parameters of PM₁₀ are presented in Table 52 and scatter plots in Figure 42. In general, model performance parameters for PM₁₀ for most models show better results than for PM_{2.5}, but are generally still low. R² results range from 0.4 (I/O ratio and MB model 1) to only 0.174 (MB model 3b). The IA ranges from 0.098 for the I/O ratio to 0.584 for the MB model 3b. Only the MB model 3b shows an IA above 0.5 indicating good model fit. NMSE are high from 7.116 (REG-J 2b) to 23.625 (MB1). Four of the simpler models underpredict the monitored (I/O ratio, REG-J 2a, MB1, MB2). The other four, more detailed models overpredict the monitored data (REG-J2b, REG-J3, MB3a, MB3b). FB ranges from 0.057 (REG-J 3b) to 0.520 (MB3b).

Model	R ²	IA	NMSE	FB
I/O Ratio	0.004*	0.098	11.898	-0.294
REG-J 2a	0.017**	0.160	11.865	-0.304
REG-J 2b	0.089**	0.343	7.116	0.119
REG-J 3	0.061**	0.282	7.833	0.057
MB1	0.004*	0.120	23.625	-0.887
MB2	0.050**	0.389	16.645	-0.381
MB3a	0.076**	0.451	9.107	0.388
MB3b	0.174**	0.584	8.108	0.520

Table 52: Model performance parameters (R², IA, NMSE, and FB) for indoor-outdoor (I/O) ratio, regression models (REG-J) and mass-balance models (MB) applied to predict PM₁₀ concentrations at Home A. R² (r squared), IA: Index of Agreement, NMSE: Normalised Mean Square Error, FB: Fractional Bias. * p < 0.05; ** p < 0.01

As for PM_{2.5}, the I/O ratio has one of the lowest performance results with an R² of only 0.004, an IA of 0.098, and a high NMSE of 11.898. The I/O ratio shows underprediction of the monitored concentrations with an FB of -0.294.

For the regression models, REG-J2a shows the lowest performance results with a low R² of 0.017, and a low IA of 0.160. The NMSE is high with 11.865 and the FB indicates underprediction with -0.304. REG-J model 2b shows a slightly better performance (R²: 0.089, IA: 0.343) and a lower NMSE of 7.116. The FB shows an overprediction of monitored concentrations with 0.119. Combining the time model results with model 2b results decreased performance for the REG-J model 3b (R²: 0.061, IA: 0.282). REG-J model 3b also shows a slightly greater error with an NMSE of 7.833, but less overprediction with an FB of 0.057. The scatter plots for the regression models show differences in predicted source related activities between REG-J model 2a and 2b. Results for regression model 2b appear generally more differentiated between different source related situations.

For the MB models, an improvement of performance with added source parameters can be observed. MB1 shows some of the lowest results for model performance (R²: 0.004, IA: 0.120) and

the highest NMSE of 23.625. The model additionally shows an underprediction by more than a factor of 2 (FB: -0.887). After including cooking source emissions in MB2, R^2 and IA show better results and the NMSE is lower (R^2 : 0.050, IA: 0.389, NMSE: 16.645, FB: -0.381). Performance was further improved with model 3a (including “active occupancy”) resulting in an R^2 of 0.076, and an IA of 0.451; the error was lower with a NMSE of 9.107. The MB model 3a shows overprediction with an FB of 0.388. MB3b (“hour after cooking” added) shows the highest R^2 of 0.174. The IA is 0.584, indicating good model fit (IA > 0.5, see chapter 3). The NMSE, with 8.108, was less than for other MB models. The FB shows a comparably high overprediction with 0.520. The scatter plots of the MB models show, as for $PM_{2.5}$, the successive addition of source activity parameters from left to right. Respective results for each source related situation show increased concentrations with the introduction of each parameter. It is however evident that the variability within each source related category is not well reflected. For “active occupancy” adding variability by minutes of source activity results in similar concentrations for many predictions at around $100\mu g/m^3$, as can be seen for scatter plots of MB3a and MB3b. Active occupancy of the indoor microenvironment often lasts a full hour. For many hours with “active occupancy” the same total value ($E_{move} * 60$) was therefore added resulting in these similar values.

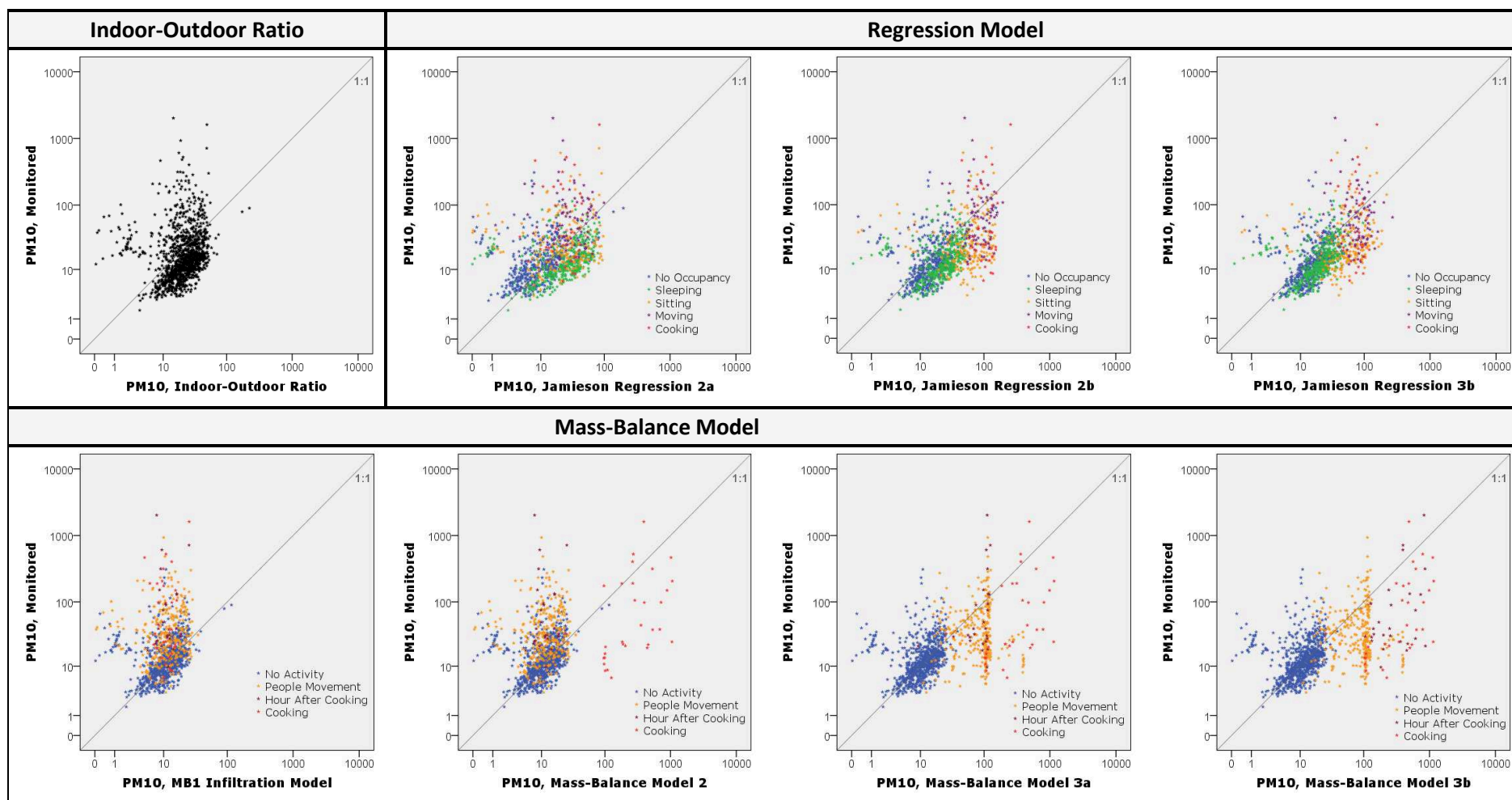


Figure 42: Modelled against monitored indoor concentrations for PM_{10} (in $\mu g/m^3$) using indoor-outdoor ratio, regression models (2a, 2b, 3), and mass-balance models (1, 2, 3a, and 3b).

Different source activities are added. All presented plots use log scales both for X and Y axes. 1:1 reference line is added to each graph.

5.3.6. Discussion

All applied models showed very weak model performance for predictions of indoor concentrations at home A. For the MB model, it could however be shown that performance increased with each added parameter of cooking, “active occupancy” and “hour after cooking”. Both for PM_{2.5} and PM₁₀ model MB3b showed the best results for model performance. For PM₁₀, MB3b model shows a good model fit with an IA above 0.5, results of other parameters however remain weak.

The generally weak performance of the results may show that all applied models for indoor particle concentrations, as compared in this analysis, have difficulties to reflect the full variability of indoor concentrations. Several other reasons may also underlie the observed weak performances:

1. The datasets for indoor concentrations both for PM_{2.5} and PM₁₀ show a positively skewed distribution, with a few concentrations (for example during cooking events) much higher than the mean. These high monitored concentrations were badly reflected by all models. For the calculation of an R² the high concentrations act as leverage points, which have a strong influence on the line of best fit. As a consequence R² results are low and error (NMSE) is high. Scatter plots however reveal a linear relationship between monitored and modelled concentrations for lower values, at a log-transformed scale.
2. Every measurement is subject to measurement error. Measurement error is the difference between the measured value and the true value (Bell 1999). Measurement error was reduced for data collected at home A by calibrating the initial measurements (undertaken using Osiris devices) to the EU reference method. A certain degree of measurement uncertainty however remains (especially for PM_{2.5}, which only qualified as indicative of reference equivalence, after calibration). A high measurement error could reduce the performance of model predictions.
3. Concentrations monitored in home A may not be representative for a typical home. Many factors influence the concentrations in a home and each of these factors can differ between homes. Unusual building characteristics or unusual behaviour of the occupants may have reduced the ability of a model to predict indoor concentrations.

Despite generally weak performance clear differences can be observed in how well the different models reflect variability between source related situations. Models that are based on outdoor concentrations only (I/O ratio and MB1 model) or include only one additional parameter (such as model REG-J2a, or model MB2) reflected little of the observed variability. Models which included more parameters for source emissions performed in comparison better. The variation between different source related situations was reflected to some extent by these more detailed models (see

Figure 41 and Figure 42). Several parameters applied for source adjustments however clearly over- or underpredicted the mean source related concentrations. In addition, these mostly static source parameters can only adjust for the variability within each source related situation to a limited degree.

Indoor/outdoor ratio

The I/O ratio shows some of the lowest model performance results and poor reflection of the distribution of monitored data. The I/O ratio remains the most applied type of indoor model for personal exposure, which raises concerns for potential introduction of exposure misclassification.

Regression Models

The regression model 2a shows very weak model performance results. The variable for occupancy, as the only second variable (apart from outdoor concentrations) does not substantially improve results compared to models based only on outdoor concentrations. The regression model 2b with differentiation between types of sources clearly performs better than model 2a. The model performance results still remain very weak for both $PM_{2.5}$ and PM_{10} and the applied coefficients do not reflect the differences between source related situations well. For example average cooking related concentrations are greatly underpredicted for both particle size fractions (see Figure 41 and Figure 42). No other model has included the differentiation between sitting and moving, which provides some added variability between different types of people's activities indoors. Including the TEMP model for regression (REG-J) model 3b did not further improve performance, which confirms weak results from the validation for the REG-J3b model, as undertaken by Jamieson (2012). Model 2b therefore remains the most convincing of the three regression models. Validation results for the model 2b, as undertaken by Jamieson (2012), showed for the fine particle size fraction a low R^2 of 0.055, which is higher than the R^2 of 0.035 observed for the application in this thesis. For PM_{10} the validation results by Jamieson (2012) showed much higher results with an R^2 of 0.559 compared to only 0.089 observed for the application in this thesis. The regression models by Jamieson (2012) showed several limitations for the application in this thesis, most notably a small dataset for model development (and validation). In addition the model for fine particles was developed for $PM_{0.5-3.5}$ not for $PM_{2.5}$. Differences between fine particle size fractions for indoor deposition and resuspension have been described in several studies (see chapter 2). The coefficients of a regression model developed with $PM_{0.5-3.5}$ therefore may not reflect $PM_{2.5}$ dynamics well. Weak model performance results shown for the application for Home A strengthen existing concerns about transferability of

the regression models. Despite future potential of this regression approach, further validation is considered necessary before the application of this approach as part of personal exposure models. In addition, further variables such as “hour after cooking” could be considered during a re-development.

Mass-Balance Models

The MB models 1, 2, 3a, and 3b perform better with each added parameter. MB model 1 is only based on outdoor concentrations and results for model performance are extremely low. In comparison to the simpler I/O ratio no improvement could be observed when using the MB1 model. Only including cooking source strength in MB model 2 increased performance, but generally model performance results remained low. The additional cooking parameter affects in addition only a few hours in the dataset (31 hours). MB model 2 represents however one of the most applied MB model types. The addition of active occupancy for MB models 3a and 3b therefore reflects indoor source situations better. The addition of an “hour after cooking” parameter further improved model performance. While the source parameters clearly improve the prediction of indoor concentration variability, the scatter plots show large over- and underpredictions of the parameters (Figure 41 and Figure 42). The cooking parameter for $PM_{2.5}$ by He et al. (2004) underpredicts observed cooking concentrations. The cooking parameter for PM_{10} from the PTEAM study (by Wallace et al. (1996)) on the other hand greatly overpredicts concentrations. The study by He et al. (2004) is small (15 homes) and may not have produced entirely representable results (especially considering the differences in emissions connected to different cooking types). The PTEAM study on the other hand collected concentrations for the study using passive sampler and calculated cooking source strength indirectly. This may have produced estimates that are not representative for the source activity. It is also possible that the cooking times recorded at home A are not typical or representative for average cooking source related concentrations. Some variability between cooking events was included by the MB model in form of the duration of source emissions. Adding variability of a source by including the temporal duration however shows limited usefulness for “active occupancy”, which often lasts all 60 minutes. The added parameter of active occupancy again clearly improved model performance, but the variability within the source related situations was not reflected. As an alternative the distinction between numbers of occupants could be considered. Data for number of occupants is often widely available even for larger population size datasets, which may make this a widely applicable option. Adding the hour after cooking in MB model 3b provided further improvement to the model. The parameter is however entirely dependent on the cooking parameter and therefore subject to the same over- and underprediction introduced for cooking. MB model 3b showed the best model

performance results and provided relatively good reflection of source related situations in comparison to the other models. Therefore later in this thesis the MB model 3b was used to calculate indoor concentrations for personal exposure model.

Little is known from the literature about the performance of MB models, as applied in the comparison (one compartment, deterministic parameters). An MB model was applied to predict indoor aerosol concentrations (a pollutant including total suspended particles and gases) by Koutrakis et al. (1992), using among others kerosene and wood burning as source parameters. A scatter plot of modelled to monitored concentrations for 28 homes was interpreted by the author to show a good correlation. In a study by Gerharz et al. (2009) an MB model is used to predict indoor PM_{2.5} concentrations in homes, including smoking and cooking as parameters. The modelled and monitored indoor concentrations have been visualised in the study in a line graph for 30 minute periods during one day. The author concluded that the sleeping period was underestimated and a period of smoking was overestimated. For the rest of the day the model showed similar concentrations compared to monitored particles.

In summary

In summary, predictions of indoor concentrations for home A resulted in generally weak results for model performance for all compared models. Variability of indoor concentrations is to a large part driven by source activities which can cause very high concentrations for a few hours. These concentration peaks have been poorly reflected by all models. Attempts have been made to predict source related indoor concentrations with mostly static parameters, which improves predictions. Emission strength of different sources are however highly variable and can therefore not be reflected fully by static parameters. Further variability could be introduced using more specific parameters (such as type of cooking or number of occupants). The development of such parameters would however require a large multi-home study, which lies beyond the scope of this thesis. For the Home A dataset it can furthermore be observed that several of the applied parameters over- and underpredict source strengths. For the regression models the over- and underprediction of indoor activities consolidated existing concerns about model transferability. The regression models were therefore not integrated into the personal exposure model, developed for this thesis. Results for the MB model also raised concerns about model performance in general, and in particular about the literature based parameters for cooking. The newly developed parameter of active occupancy suffers from its static approach, but still improved model performance. The newly developed

addition of hour after cooking further improved performance, but its value is dependent on the (over- and underpredicting) cooking parameters.

For the application as part of the personal exposure model the MB3b model was used to predict indoor concentrations. As a comparison, the I/O ratio was applied in a modified model of the personal exposure model. Applying both the MB model 3b and the I/O ratio provides the possibility to compare the extent of exposure misclassification when different indoor models are used as part of personal exposure modelling.

5.4. Modelling Office Indoor Exposure

5.4.1. Introduction

About 30% of time spent indoors is spent at work (Schneider et al. 2004). Personal exposure models have therefore commonly included the indoor work ME. For this purpose some studies have used an indoor to outdoor ratio developed for home indoor MEs (Dons, Van Poppel, Kochan, et al. 2014; Gariazzo et al. 2014). Modern indoor work microenvironments are however mostly mechanically ventilated and there are usually few indoor sources present. For these reasons offices show generally lower concentrations compared to ambient concentrations, as well as compared to homes indoors (Hänninen 2005; Ho et al. 2004; Gupta & Cheong 2007; Horemans et al. 2008; Ní Riain et al. 2003). It is therefore important to adjust office concentrations from surrounding ambient concentrations to avoid overprediction. An introduction to the literature on mechanically ventilated offices has been presented in chapter 2. In summary, studies on mechanical ventilation show that the ventilation systems reduce particles through filtration. A further particle reduction is often achieved because many ventilation systems draw the incoming air from a high position, such as roof level. At this height concentrations are much lower than close to the ground. In order to adjust concentrations from the ambient to reflect reduced in-office exposure specific adjustments are selected from the literature in the following paragraph.

5.4.2. Adjusting for the Indoor Office Microenvironment

By far the biggest study that looked at the infiltration of PM_{2.5} into mechanically ventilated indoor work spaces has been undertaken by Hänninen et al. (2005) including 94 buildings in Finland. Hänninen et al. (2005) calculated a model based on an infiltration factor (see Equation 2) and calculated indoor work place exposure (C_i) as follows:

Equation 16: PM_{2.5} indoor office model

$$C_i = F_{inf} \times C_a + \sum_j C_{sj},$$

Where *F_{inf}* is the infiltration factor, *C_a* the ambient PM_{2.5} concentration, and *C_{sj}* represents possible source activities within the microenvironment. The office infiltration factor (*F_{inf}*) for PM_{2.5} calculated by Hänninen et al. (2005) is 0.35 and without source activity effectively can be applied in the same way as an I/O ratio. However, two limitations apply: 1) general work places (not only offices) were included and 2) mechanical ventilation was only assumed by age of building (post-1990 offices were assumed to have mechanical ventilation). Another study including two offices was conducted in Hong Kong and showed a slightly higher average I/O ratio for PM_{2.5} of 0.5 (Ho et al. 2004). A third study included five offices in Belgium and showed I/O ratios for PM_{2.5} of between 0.34 and 0.77 (Horemans et al. 2008). All of these I/O ratios lie below the home I/O ratio from the EXPOLIS study (Koistinen et al. 2001) of 0.86 applied above, as well as below most other home I/O ratios presented in chapter 2. Despite its limitations the infiltration factor calculated by Hänninen et al. (2005) was considered the most informed estimate for outdoor to indoor particle transfer for PM_{2.5}. The infiltration of 0.35 was therefore used to estimate office concentrations for this study. Offices monitored for this thesis showed few source activities (see chapter 3). Source activities were therefore not included. Subsequently the infiltration factor could simply be multiplied by the outdoor PM_{2.5} concentration similar to an I/O ratio.

For PM₁₀ very few results on office MEs are available from the literature. Only one study, by Gupta & Cheong (2007) presents indoor and outdoor results for fully mechanically ventilated office buildings. Limitations of this study are that 1) the measurements were undertaken in Singapore (providing a different general environment to the UK) and that 2) the study included only two offices. An I/O ratio of 0.46 was calculated by this study. This estimate lies below the home I/O ratio for PM₁₀ of 1 from the PTEAM study (Özkaynak et al. 1996) used for home I/O ratios above. The I/O ratio of 0.46 was considered the best available estimate for in-office adjustments and was therefore applied to calculate PM₁₀ office concentrations for the personal exposure model.

5.4.3. Conclusions

The main aim for this section was to find suitable adjustments for mechanically ventilated office MEs to avoid overprediction of concentrations for hours spent at work. The selected indoor to outdoor functions clearly reduce ambient concentrations for the ME, and remain well below established I/O ratios for home MEs. The estimates show however some limitations of data quality for PM_{2.5} and

data quantity for PM₁₀. In-office adjustments selected above are applied in the personal exposure model and results for their application are evaluated in chapter 6.

5.5. Chapter Summary

In this chapter a set of ME adjustments, models and estimates was prepared for the integration into the personal exposure model in chapter 6.

First, adjustments of transport MEs for walking, cycling and in-bus exposure were calculated from data collected during a bespoke transport monitoring campaign (see chapter 3). Furthermore, adjustments and estimates for in-car and in-railway transport were reviewed from the literature.

Modelling approaches for home indoor MEs were compared in a second section of this chapter, revealing generally weak performance by most models. The best performance was observed for an MB model including two new parameters, which were calculated as part of this thesis: active occupancy and hour after cooking.

Furthermore, an adjustment ratio for exposure in fully mechanically ventilated offices was selected from the literature for application in the personal exposure model.

All different types of MEs and how they are modified for the personal exposure model are shown in Table 53. Outdoor sitting, standing and walking MEs are calculated directly from the model for ambient concentrations (see chapter 4). For all other MEs, modifications are undertaken, as described above. Office indoor, cycling, in-car, and in-bus MEs are adjusted by using a ratios, for times during rail transport estimate from the literature are applied. The home indoor ME, as well as ‘other indoor’ MEs are calculated using the mass-balance (MB3b) model described above.

MEs	ME model	PM _{2.5} Ratio (Estimate)	PM ₁₀ Ratio (Estimate)
Outdoor sitting or standing	-	-	-
Outdoor walking	-	-	-
Home Indoor	MB model	For parameters, see above	For parameters, see above
Office Indoor	Office I/O ratio	0.35	0.46
Other indoor*	MB model	For parameters, see above	For parameters, see above
Cycling	In-Traffic ratio	1.39	1.32
In-Bus	In-Bus ratio	1.65	2.57
In-Car	In-Car ratio	1.46	1.42
In-Train	Literature estimate	(20.64)	(183.41)

Table 53: Exposure microenvironments integrated into the MEPEX model, as well as type of model or estimate applied for microenvironmental adjustment.* naturally ventilated

Exposure during cycling is modelled using the ambient LUR-adj model to calculate hourly exposure along the route, which is then adjusted to model inside traffic by the in-traffic ratio of 1.39 for PM_{2.5} and 1.32 for PM₁₀. In-bus exposure is similarly calculated first for the hourly exposure along the route and then adjusted to represent in-bus exposure with the in-bus ratios of 1.65 for PM_{2.5} and 2.57 for PM₁₀. The estimates for in-train exposure are 20.64µg/m³ for PM_{2.5} and 183.41µg/m³ for PM₁₀. For home locations the MB model is applied, including source contributions from people's active occupancy, cooking and for the hour after cooking. For the office indoor ME the in-office I/O ratio of 0.35 for PM_{2.5} and 0.46 for PM₁₀, is applied.

'Other indoor' MEs (not home or work) are also modelled using the MB model, as evidence for specific models is sparse for most other indoor MEs (Gariazzo et al. 2014) with the possible exception of schools. Applications in the context of this thesis however focus on adult populations, an in-school model or adjustment could be added if required for future applications. Most 'other indoor' MEs are public or community spaces (bars, sport centres etc.), which generally include occupants. The application of the MB model for these spaces gives the option to include people's occupancy, as well as possible further sources (such as cooking when visiting restaurant). The application of the MB model (as used in this context) for other indoor MEs is however limited to naturally ventilated indoor MEs.

An additional modified personal exposure model is composed in chapter 6, for which the calculation of personal exposure is based on PM concentrations at the home location for all MEs. Home locations are mostly located in areas with background concentrations, adjustments for in-transport from background concentrations were therefore preferred for this model (in contrast to in-transport to roadside adjustments, presented above). In-transport to background ratios have been discussed above and are summarised in Table 54.

MEs	PM _{2.5} Ratio (Estimate)	PM ₁₀ Ratio (Estimate)
Walking	1.27	2.13
Cycling	2.01	2.41
In-Bus	2.07	4.14
In-Car	1.57	1.44
In-Train	(20.64)	(183.41)

Table 54: Transport exposure microenvironments integrated into the MEPEX-res model, as well as the applied ratio estimate applied for microenvironmental adjustment.

For walking the mean ratios for both the low traffic and high traffic routes developed from concentrations of the transport monitoring campaign are applied. For cycling, as well as for in-bus exposure, the mean ratios calculated for the high traffic routes are applied. Ratios calculated by

Zuurbier et al. (2010) are applied for in-car adjustments. The ratios calculated for diesel cars by Zuurbier et al. (2010) are preferred over ratios presented for gasoline cars, as more licenses in Britain are issued for diesel cars, compared to gasoline cars (Department for Transport 2013).

6. Building and Evaluating Models of Personal Exposure

6.1. Introduction

This chapter has three main aims, the first aim is to build an integrated personal exposure model based on the components selected and developed in chapter 4 and 5. The second aim is to evaluate performance of the model by applying it to monitored personal exposure. The third aim is to compare predictions with estimates from other approaches for personal exposure modelling.

In chapter 5 all the components for the construction of a personal exposure model were selected or developed: 1) A model for ambient concentrations was developed which provides predictions both temporally and spatially at a high resolution. 2) Adjustments for exposure in different transport modes were calculated or selected from the literature. 3) The mass-balance modelling approach was selected as a model for home indoor concentrations, and 4) an in-office adjustment was chosen to predict exposure during times at work. These components are integrated in this chapter to the MicroEnvironmental Personal EXposure (MEPEX) model.

Most existing personal exposure models have in contrast used simpler approaches, for example using indoor-outdoor (I/O) ratios to predict home indoor concentrations or assuming at-home location throughout the day. It was therefore important to understand how simpler approaches, as applied in the literature compare to the MEPEX model. Modified variations of the MEPEX model were therefore built, based on different approaches in the literature. Model performance of the MEPEX model and the modified model variations were then evaluated by applying all approaches to predict hourly and daily personal exposure for individual A (see chapter 3). Additionally, simple exposure estimates based on home ambient concentrations are used as a comparison. Predictions by the different approaches are then compared. Strengths and weaknesses of the different prediction methods are discussed. The model variations give different importance to model components. Comparing the model variations should therefore additionally reveal information about the importance of the components.

Studies using personal exposure models have been introduced in chapter 2. In Table 55, these studies are presented, detailing different approaches and the components they use to predict personal exposure. The main differences between the studies are:

- Type of model for ambient concentrations,

- Geographic locations for times away from home,
- Included MEs,
- Model type for home indoor MEs,
- Model type for work indoor MEs,

For most of the studies ambient concentrations underlie the calculations of concentrations in all different MEs. Some studies however did not differentiate between spatial locations for each individual, and directly applied ME models and adjustments to an estimate of home ambient concentrations (Borrego et al. 2009; Gerharz et al. 2009). The MEs included in the personal exposure models vary, with most models specifying home indoor and work (or school) indoor, as well as outdoors. Modelling of the home indoor ME also differs between studies: Most studies use an I/O ratio or I/O regression function, only the EXPOLIS study, as well as a study by Gerharz et al. (2009) use an mass-balance (MB) model for predictions. For the work ME, some studies applied specific I/O ratios calculated for the work ME. Other studies however applied the methods calculated using home indoor concentrations to predict exposure at work.

Some of the main questions these different approaches raise are: Is there a difference between predictions based only on ambient concentrations at all visited geographic locations, compared to predictions assuming home ambient concentrations for the whole day? What is the impact on personal exposure predictions if a simple adjustment, such as an I/O ratio is used to predict concentrations at home indoors instead of using an MB model? What is the impact on personal exposure predictions if an I/O ratio based on measurements at home is applied to predict in-office exposure instead of using an I/O ratio calculated with concentrations monitored in offices?

Apart from approaches used for personal exposure models, the many exposure predictions in epidemiological studies use concentrations from network monitoring stations as a proxy for home ambient concentrations. An additional question was therefore: How well can concentrations at monitoring stations (nearest station or an average of stations) predict personal exposure and how important is the addition of ME adjustments for the predictions?

<i>Study</i>	<i>Pollutant</i>	<i>Location</i>	<i>Individual time-space patterns</i>	<i>MEs</i>	<i>Home Indoor Model</i>	<i>Work (or School) Indoor Model</i>	<i>Adjustments for in-traffic MEs</i>
AB²C (Dons, Van Poppel, Kochan, et al. 2014)	BC	Flanders, Belgium	Yes	Home, in-traffic (motor or active), work	I/O ratio	Home I/O ratio	Estimates (FEATHERS model)
Borrego et al. (2006)	PM ₁₀	Lisbon, Portugal	Yes	Home indoor, office indoor school indoor, in-transport	I/O regression function	I/O regression function for offices	In-vehicle regression function (Gulliver & Briggs 2004)
Borrego et al. (2009)	PM ₁₀ (O3)	Portugal	No (area)	Indoor places, outdoor	I/O ratio	I/O ratio for work/school	none
EXPAND (Baklanov et al. 2007; Hänninen 2005b; Kousa, Kukkonen, et al. 2002)	PM _{2.5} (NO2)	Helsinki, Finland	Yes	Outdoor, indoor, other	I/O ratio	I/O ratio (not specified)	None
EXPAH (Gariazzo et al. 2014)	PM _{2.5} , (PAH)	Rome, Italy	No (residential areas)	Outdoor, home, school	I/O ratio	Home I/O ratio	Bus & car: Considered equal to outdoor, underground: home I/O ratio
EXPOLIS (Kruize et al. 2003; Hänninen et al. 2003)	PM _{2.5} , PM ₁₀	Netherlands, Athens (Greece), Basel (Switzerland), Helsinki (Finland), and Prague (Czech Republic)	No	Outdoor, home indoor, work indoor, other indoor, in-transport	Sampled from distribution of MB model or monitored	Sampled from distribution of MB model or monitored	Approach 1: random sample from central monitoring station. Approach B: sampled from MB modelled distribution for homes
Gerharz et al. (2009)	PM _{2.5} , PM ₁₀	Munster, Germany	Yes	Outdoor, home indoor, work indoor, car, bus	MB	MB for non-mech. Ventilated office/university	Ratios for in-car and in-bus
STEMS (Briggs et al. 2005; Gulliver & Briggs 2005)	PM ₁₀	Northampton, UK	Yes	tbc	tbc	tbc	In-vehicle ratio

Table 55: Integrated personal exposure models, as developed by European studies, and their model components. PAH = Polycyclic aromatic hydrocarbons, tbc = to be confirmed.

In this chapter the questions summarised above are explored, comparing predictions for monitored personal exposure between different methods. In the next section, the different modelling approaches and their development are described. Then, predictions by the MEPEX model for each main ME visited by individual A are presented. This is followed by the evaluation of personal exposure by all different methods for individual A. In order to examine if the MEPEX model also predicts similarly well for exposure of other individuals (apart from individual A), the MEPEX model is applied to model personal exposure concentrations monitored by individuals B, C, and D. A model application to a study population in London using simplified virtual time-activities is in addition undertaken to demonstrate model applicability to a larger population.

6.2. Introductions and Preparation of Applied Methods for Personal Exposure Modelling

In this section the personal exposure model, MEPEX, as well as all other methods used to predict personal exposure are described. Furthermore, data preparation for the analysis is presented.

6.2.1. The MicroEnvironmental Personal EXposure (MEPEX) Model

The MEPEX model predicts personal exposure by time-weighting exposure for different MEs visited during the day. This general approach has been used by other studies. Few other models have however been developed which integrate both a model for ambient concentrations of high temporal and spatial accuracy, as well as detailed adjustments for MEs.

The MEPEX model is constructed from the following components:

- Temporally and spatially refined ambient concentrations
- Time-activity of individuals
- ME models, ratios and estimates

As a first step to calculate the MEPEX model, ambient concentrations for all locations visited by the modelled individuals are calculated using the ESCAPE-LUR model. For the application temporal adjustments, concentrations from network monitoring stations in the area are averaged and applied as an adjustment to the LUR predictions (see chapter 4). ME models and adjustments are then applied to the spatially and temporally refined ambient predictions according to space-time-activity patterns. Calculations of hourly exposures were undertaken by first calculating predictions for all MEs separately, then these estimates were time-weighted for minutes per hour spent in each

different ME. The hourly estimates were then aggregated into daily exposure predictions. All different types of ME modifications have been discussed in chapter 5 and are summarised in Table 53.

Application of the MEPEX model to individuals A, B, C, and D

The MEPEX model was applied to all individuals from the personal monitoring campaign (A, B, C, and D). Modelling of home MEs included the subject's own homes, as well as one home of a friend. Home volumes were required as MB model input and were provided by the occupants: 157.5m³ for Home A, 40.50m³ for Home B, 150m³ for Home C, 175m³ for Home D, and 200 m³ for the friend's home. Two other indoor MEs were visited during the monitoring campaigns: a nursery (2 visits, 35min, individual C) and a doctor's practice (1 visit, 38min, individual A). Both spaces were naturally ventilated and the MB model was applied. People's active occupancy was assumed for the time of the stay. No accurate data on the volume of these two indoor spaces was available. The volume of both spaces was therefore estimated by the visiting individual at 200m³ each.

6.2.2. Alternative Methods of Personal Exposure Assessment

The performance of the MEPEX model was compared to estimates using ambient concentrations, as well as modified model versions of the MEPEX model. All methods were subsequently calculated to predict concentrations for individual A and results between the model predictions were compared for hourly and daily estimates. The methods are described in the following paragraphs and an overview of their components is given in Table 56.

As mentioned before, many existing studies base the exposure estimates of a population directly on monitored concentrations using data from monitoring stations as approximations for outdoor concentrations at home. In order to test the performance of this method in comparison with more informed estimates for personal exposure, PM_{2.5} and PM₁₀ concentrations from the stations nearest to the home, as well as for the average concentration of monitoring stations in the study area, are applied here to predict concentrations collected during personal monitoring. Predicting personal exposure using concentrations from the monitoring station average is referred to as the AVERAGE method. Predictions using the nearest monitor are referred to as the NEAR method. Details about the monitoring station data used for this application are presented under "Data Preparation", below. Models AMB-h and AMB-ta are both based only on ambient concentration predictions of the LUR-adj model. The LUR-adj model uses predictions for the home outdoor concentrations for all times,

the LUR-ta model uses all spatial locations as defined by the time-activity of the individual to calculate estimates for personal exposure. The main purpose of the AMB-h model here is to evaluate the difference to the AMB-ta model and as such evaluate the importance of using spatially differentiated predictions. The temporal variability of the AMB-h model is however the same as for the AVERAGE method. Comparing the AMB-ta model with the MEPEX model in addition examines the impact of including ME models into the predictions.

Methods	Ambient Prediction	Spatial Differences	Time -activity	ME models
AVERAGE	Monitoring data	-	-	-
NEAR	Monitoring data	-	-	-
AMB-h	LUR-adj	-	-	-
AMB-ta	LUR-adj	√	√	-
ME-h	LUR-adj	√	√	(√) I/O ratio for homes
ME-o	LUR-adj	√	√	(√) home I/O ratio for office
MEPEX-res	LUR-adj	-	√	√
MEPEX	LUR-adj	√	√	√

Table 56: Approaches of personal exposure predictions and model components they include.

Exposure models ME-h and ME-o compare the impact of different ME models to the personal exposure model. Model ME-h looks at the application of a home I/O ratio instead of the MB model. The I/O ratio has been shown in chapter 5 to predict home indoor concentrations less well than the MB model. Yet, I/O ratios are widely applied in personal exposure studies. The impact between using the MB model or the I/O ratio to personal exposure estimations is therefore examined here. Model ME-o changes the office I/O ratio into the home I/O ratio. Only very few studies have investigated exposure to particles in modern office MEs, in several studies I/O ratios developed for indoor home MEs have therefore been applied to estimate concentrations in office MEs (Gariazzo et al. 2014; Dons, Van Poppel, Kochan, et al. 2014). The difference between these approaches is tested here by comparing model ME-o to the MEPEX model.

The MEPEX-res model included all types of ME models (traffic adjustment, MB model), but is based on ambient concentration for the home location, assuming location at the residential address for all visited MEs. Limited or no spatial differences during time-activities have been assumed by several personal exposure studies (Kruize et al. 2003; Hänninen et al. 2003; Gariazzo et al. 2014; Borrego et al. 2009). Predicting ambient concentrations only for residential addresses of a study population can significantly simplify the calculations involved in personal exposure predictions. The comparison of the MEPEX-res model with the MEPEX model provides an evaluation of this more simplistic, yet more transferable approach.

Ambient concentration predictions for home addresses, as underlying the MEPEX-res model, reflect background concentration levels at most home locations. Adjustments for in-traffic modes made for the MEPEX model are however based on roadside predictions of ambient particle concentrations. For the application of the MEPEX-res model different in-transport adjustments were therefore needed. Background to in-transport adjustments, as reviewed or calculated in chapter 5 are therefore applied. A summary of these ratios is given in Table 54. All methods were subsequently calculated to predict exposure for individual A and model performance was compared for hourly and daily estimates.

6.2.3. Data Preparation for Analyses

Personal Exposure Monitoring Data

For the evaluation and comparison of the personal exposure model, data from the personal exposure monitoring campaigns was used. The concentrations monitored during these campaigns have been described in detail in chapter 3. Personal monitoring had been undertaken by individual A during 43 days. Individuals B, C, and D monitored their personal exposure during between 2 and 4 days. Due to the much larger dataset, personal monitoring data of individual A is used for detailed evaluation and comparison of the modelled concentrations in this chapter. In order to assess transferability of the model to different individuals, the MEPEX model is then applied to predict concentrations of the other three individuals.

In preparation for the application, the personal monitoring data was aggregated to hourly estimates. Several hours showed less than 60 minutes of concentrations. Missing data points in a dataset can potentially cause limited representativeness for the examined time period. This occurs for example if one season is missing in an annual dataset, which may make the data not representative for the year. For the current analyses the representativeness of each minute in an hour is however of comparably minor importance as differences of personal exposure throughout the day are rather related to time-activities and MEs, than to 60 minute intervals. Single minute measurements can on the other hand include measurement errors or could be increased by unusual events for a situation, such as one truck passing in an otherwise quiet road. To avoid single data points to having a high influence on the hourly average, hours with very few minutes (<10) were excluded. In consequence seven hours were removed from the dataset for individual A, one hour was removed from the dataset of individual C, and two hours were removed from the dataset of individual D. For aggregation to daily estimates the hourly concentrations were furthermore weighted by number of minutes. For daily estimate a balanced representation of hours in the day is important, as the daily

routine is reflected in the numbers of hours spend in each ME. For the daily estimates of individual A, seven days were therefore removed for days with less than 13 hours of monitoring concentrations. 10 further days (out of 36) showed between 13 and 20 hours of monitored concentrations. All of these included monitoring times at home, during commute, as well as in the office and were therefore considered broadly representative of daily exposure. For individuals B and D all days included at least 23 hours. The monitored exposure of individual C however was limited to times away from home and included therefore only eight hours for each day, as well as no representative distribution of MEs. As a result, the data collected by individual C is considered of limited representativeness for personal exposure, the monitoring concentrations were only included in the graphs (for hours per ME, and for hours and days per individual), but not in the calculation of model performance parameters.

Concentrations at a seasonal level are additionally presented for personal exposure of individual A. Seasons were categorised according to meteorological categories, as described by the Met office (<http://www.metoffice.gov.uk/learning/learn-about-the-weather/how-weather-works/when-does-spring-start>, accessed on: 13th April 2014). A season includes about 90 days, the number of monitored days per season was in comparison considered low (winter: 9, spring: 8, summer: 17, autumn: 9) and unrepresentative. Seasonal averages are therefore visually presented, but seasonal performance of the prediction methods is not further evaluated.

Temporal Adjustment of LUR Model Predictions

Personal exposure monitoring was undertaken between April 2010 and September 2011 and temporal adjustment of LUR predictions was calculated according to hours of monitoring. In order to temporally adjust the LUR ambient model, hourly background average concentrations were applied, as described in chapter 4. Background monitoring stations providing data for 2010 were presented in chapter 4. Data availability at the background monitoring stations was however different for times of personal exposure modelling and data collection discontinued at several monitoring stations at the end of 2010. Background stations were therefore selected specifically to match data availability of times of personal exposure modelling. Background stations with data availability of above 50% for the relevant hours of personal exposure modelling were selected for the background average. 50% of data availability was chosen as a limit (as compared to 75% for the analysis of temporal adjustments in chapter 4) to provide a wider data base, particularly for 2011 for which generally fewer stations were available. The spatial limit for selection was set as before at the M25 for PM_{2.5} and at the London North and South Circular Road (see chapter 4). Selected monitoring stations

included stations introduced in chapter 4, as well as one additional station for PM_{2.5}: Bexley - Belvedere and three additional stations for PM₁₀: Islington Arsenal, Lambeth - Streatham Green, and Westminster - Horseferry Road. Basic statistics of all background station data for the hours of personal exposure modelling are presented in Appendix J.

Monitoring stations used for the AVERAGE and NEAR method

For the methods AVERAGE and NEAR, concentrations from monitoring stations are used to predict personal exposure for individual A. Most studies use concentrations from background stations (excluding roadside or industrial stations), which are generally considered more representative of exposure at home locations (Vineis et al. 2006). As this application attempts to reproduce the method from the literature, background stations are used both for the nearest, as well as for the station average (using LAQN definitions for 'urban background' as station type). For the average of background stations, the same stations selected for the LUR model adjustment were used, see above. For the NEAR method the nearest of those background stations from the home location was identified. For PM_{2.5} the nearest station was Camden – Bloomsbury. Missing concentrations for the dataset were filled in using data from the second nearest station (Kensington and Chelsea – North Kensington). For PM₁₀, the nearest background station was City of London - Sir John Cass School, missing data was filled in using again concentrations from the second nearest station: Camden – Bloomsbury.

Methods to Evaluate Model Performance

Model performance is evaluated by applying model performance parameters. Model performance parameters have been introduced in chapter 3 and they include R^2 , IA, NMSE, and FB. In addition scatter plots are presented between monitored and modelled concentrations.

6.3. Exposure Predictions by Microenvironment

As a first step towards calculating the final personal exposure model all MEs were calculated separately by the MEPEX model. In the following paragraphs modelling results for main repeatedly visited MEs by individual A are presented: home, office, in-bus, and cycling. Results are then compared to the monitored concentrations for the same times. Examining these MEs separately from the final model can show the specific strength and weaknesses of predictions for each ME model or adjustment before the MEs are averaged to hourly personal exposure. Only MEs are

presented here for which sufficient data was available to focus on significant results. ME results for home and office MEs are summarised by hour. As for the final model, hours with less than 10 minutes have been removed from the dataset for this analysis. Cycling and in-bus MEs are summarised by each commute instead of hours, as one single commute often took place over two hours.

6.3.1. Home

MB modelled home indoor concentrations for home A have been examined in detail in chapter 5, where results were compared to other models predicting indoor particle concentrations. For the indoor model comparison in chapter 5, monitored home outdoor concentrations were used for the calculation. Here, in preparation of the final model home indoor concentrations were calculated based on LUR-adj predictions for home outdoors. The model performance using modelled outdoor concentrations is here investigated and compared to results by the MB model using monitored outdoor concentration. This comparison can evaluate to what extent using modelled outdoor concentration as an input to the indoor model changes results.

438 hourly concentrations were examined for indoor home predictions (21 hours were removed as they included less than 10 minutes of data). Results of model performance parameters are presented together with the scatter plots in Figure 43. For $PM_{2.5}$ the results in this analysis show a generally weak performance with an R^2 of 0.060, an IA of 0.239, and a high NMSE of 8.119. The results are however similar to MB model results based on home outdoor monitoring concentrations ($R^2 = 0.051$, IA = 0.251, NMSE = 9.107). The FB observed for this analysis is 0.097 which indicates a slight overprediction of the data, whereas at the comparison using outdoor monitoring data, the FB was slightly negative. Scatter plots in Figure 43 (note: both X and Y axes are presented in a log scale) show a clear distinction of the different situations included in the model (no source, people's movement, hour after cooking, and cooking) for both particle size fractions. Situations for the scatter plots were categorised hierarchically, as in chapter 5: cooking or hour after cooking > people moving > no source. The graph shows that times with no source activities are centred around the 1:1 reference line, while people's movement (or active occupancy) appears to be overpredicted by the model for both particle size fractions. Hours of cooking and after cooking exposure are for a few concentrations considerably under- and for others considerably overpredicted. The two highest monitored hourly concentrations show particular underprediction. Removing these two values improved model performance (see model performance results in brackets, Figure 43) resulting in an IA of 0.522. This shows that the final $PM_{2.5}$ indoor model performs well for the majority of data

points. It however fails to predict some of the most important high concentrations well. The extreme underprediction of those two data points however reduced overprediction of the dataset. Removing the two data points increases the FB to 0.294.

For PM_{10} the R^2 was 0.139, which is a little lower than the R^2 of 0.174 observed for the home indoor model comparison in chapter 5. The IA of 0.543 implies good model fit and is only slightly below the IA of 0.584 calculated for the indoor model comparison in chapter 5. The NMSE for the current analysis is, with a value of 6.095, high, but below the 8.108 observed before in chapter 5. The FB is 0.633 showing overall a considerable overprediction by the model which is a little above the previously observed overprediction of 0.520.

For PM_{10} the scatter plot (Note: both X and Y axes are presented in a log scale) again shows times with no source activity predicted close to the reference line. Hours of all source related exposure situations appear however to be mostly overpredicted, which is reflected in the relatively high FB of the model.

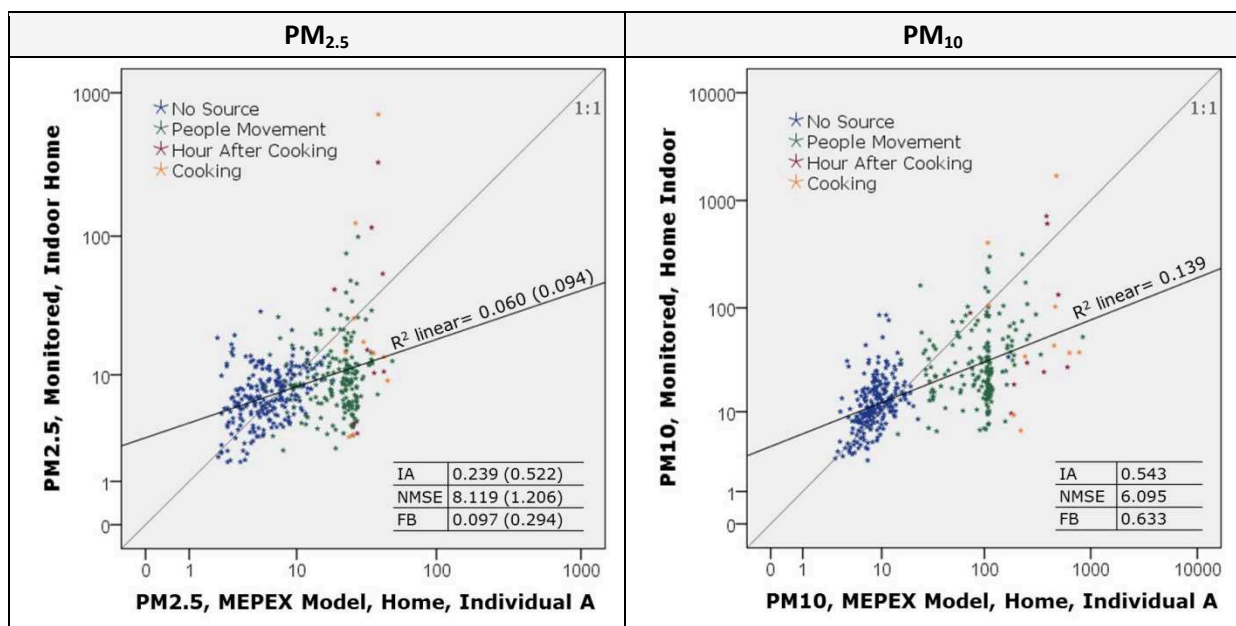


Figure 43: Modelled (MEPEX model) against monitored $PM_{2.5}$ and PM_{10} (both in $\mu g/m^3$) for home indoor concentrations collected by individual A, specified by different indoor situations. X and Y axes are presented in log scale. 1:1 reference line and line of best fit (R^2) are added. Model performance parameters are as well presented in the graph. For $PM_{2.5}$ model performance results after removal of the two highest monitored concentrations are added in brackets. Both results for R^2 are significant at a 0.01 level

In summary, the results of the final indoor model presented here show a weak model performance for $PM_{2.5}$. Removing the two highest concentrations shows that the model performs well ($IA > 0.5$) for the majority of data points. The model performance parameters for PM_{10} show a good model fit for PM_{10} ($IA > 0.5$). The R^2 of the PM_{10} results is low however, with 0.139, and the NMSE is high at 6.095. Results for performance parameters of the final indoor model both for $PM_{2.5}$ and PM_{10} are similar compared to the performance of the model calculated with monitored outdoor concentrations. The application of modelled instead of monitored outdoor concentrations only slightly lowered model performance for R^2 and IA results. The NMSE on the other hand showed lower scatter for both $PM_{2.5}$ and PM_{10} when using modelled outdoor concentrations.

6.3.2. Office

Mechanically ventilated indoor office MEs have not been examined much in the literature and the applied I/O ratios from the literature also showed some clear limitations (see chapter 5), it was therefore important to examine the performance of the applied ratios.

The analysis for office MEs visited by individual A was undertaken here for 268 hours in the office (11 hours were removed, as they covered less than 10 minutes). Model performance parameters are presented in Figure 44 together with the scatter plots. The results for $PM_{2.5}$ show a moderate R^2 of 0.375, and an IA of 0.524, indicating a good model fit ($IA > 0.5$). Yet, the NMSE is comparably high with 2.860 and the FB shows an overprediction by much more than a factor of 2 (overprediction by a factor of 2 at $FB=0.67$) with 1.068. For PM_{10} the R^2 shows a low result of 0.188, the IA is just below 0.5 with 0.472. The NMSE is relatively high with 1.609 and the FB shows, as for $PM_{2.5}$, a considerable overprediction of the model of 0.911. Scatter plots of modelled against monitored concentrations confirm the overprediction indicated by the FB results.

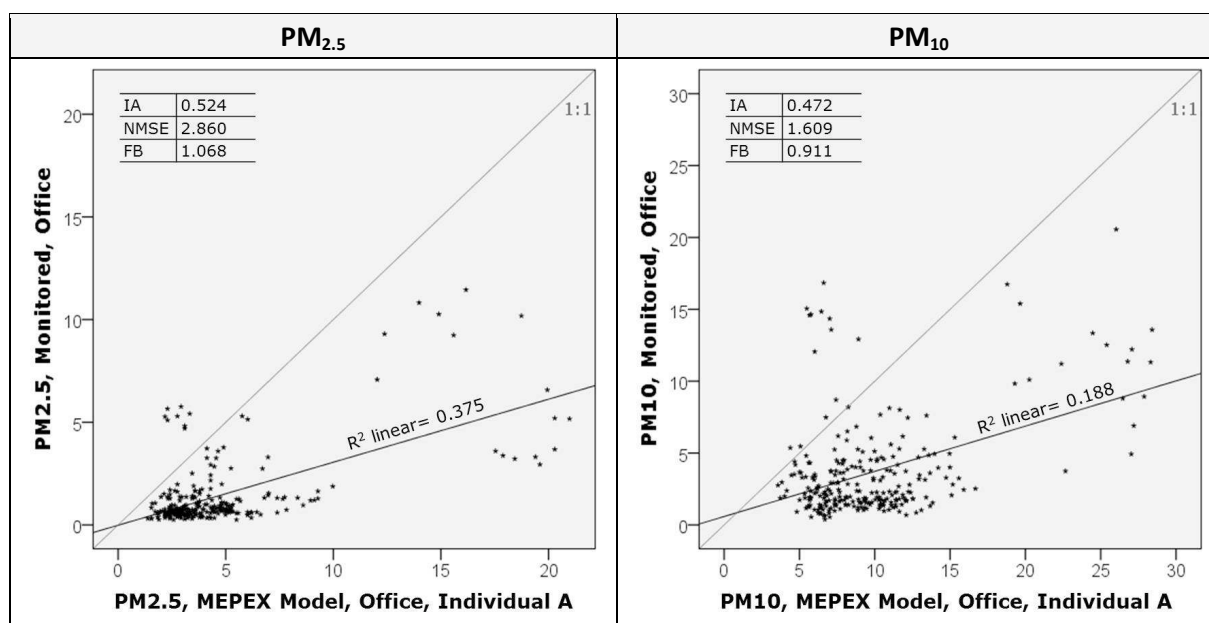


Figure 44: Modelled (MEPEX model) concentrations of hourly PM_{2.5} and PM₁₀ (both in $\mu\text{g}/\text{m}^3$) in the office microenvironment against monitored concentrations of individual A. 1:1 reference line and line of best fit (R^2) are added. Model performance parameters are as well presented in the graph. Both R^2 are significant at a 0.01 level

In summary, the results for R^2 and IA show that the variability in a mechanically ventilated office MEs can to some extent be predicted by using adjusted ambient concentrations. The adjustments taken from the literature however overpredict concentrations considerably for both PM_{2.5} and PM₁₀. The average ratio between LUR-adj predicted ambient and in-office concentrations for this office was calculated as a comparison from the monitored personal exposure used in this application. The resulting ratios are 0.14 for PM_{2.5} and 0.19 for PM₁₀, which suggests a much lower adjustment ratio should be considered for future applications.

6.3.3. Cycling Commute

47 commutes were undertaken by individual A with a bicycle. As described in detail in chapter 3 the model for the commute route was calculated from mean LUR-adj predictions along the route and adjusted for in-traffic concentrations by ratios adjusting for the horizontal displacement between sidewalk and position in-traffic. In-traffic adjustments for cycling exposure (in comparison to exposure along the sidewalk) have never been attempted in this form.

The results for model performance parameters are presented with the scatter plots in Figure 45. For PM_{2.5} the results show a high R^2 of 0.852, good model fit with an IA of 0.736, and an NMSE of 1.224. The FB shows an underprediction of -0.372. For PM₁₀ the R^2 shows a moderate result of 0.486, the IA

shows good performance of 0.578, and the NMSE equals 1.117. The FB shows a considerable underprediction of the model with -0.661 (FB of -0.67 signifies an underprediction by a factor of 2).

The scatter plots confirm the underprediction of the model both for $PM_{2.5}$ and PM_{10} . The data points in the graph are differentiated by morning and evening commute. No clear pattern between the times of the day can be detected, the highest monitored concentrations (especially for PM_{10}) have however been observed during the morning commute.

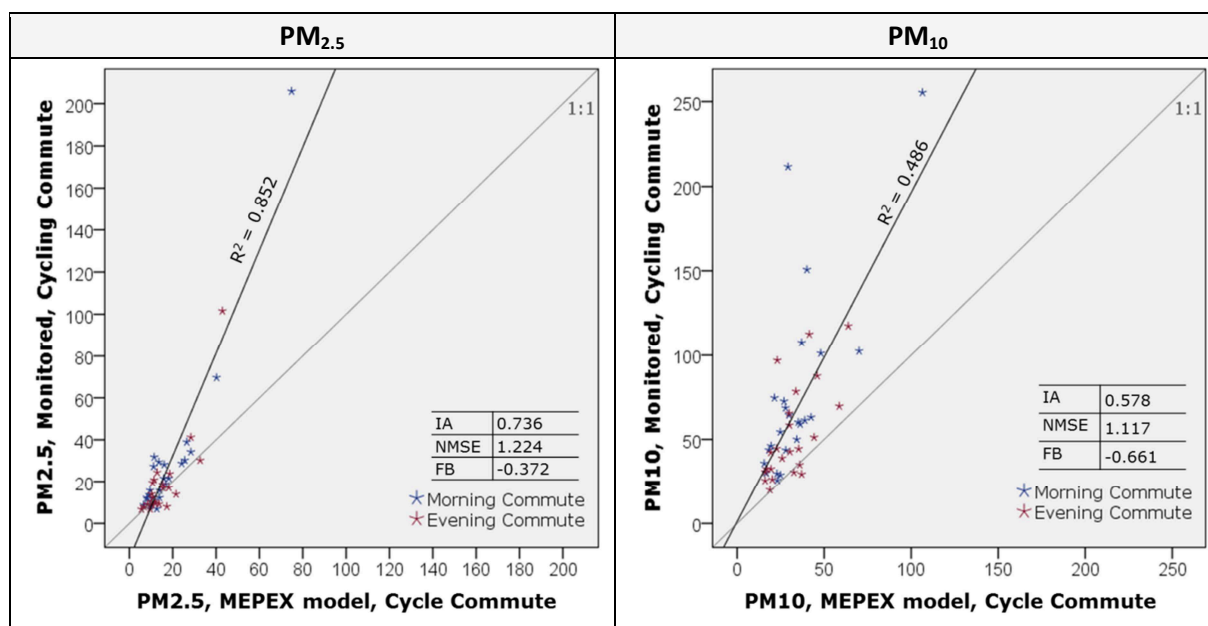


Figure 45: Modelled (MEPEX model) concentrations of hourly $PM_{2.5}$ and PM_{10} (both in $\mu g/m^3$) for cycling commutes against monitored concentrations of individual A. 1:1 reference line and line of best fit (R^2) are added. Model performance parameters are as well presented in the graph. Both R^2 results are significant at a 0.01 level.

In summary, the predictions per commute showed a good model performance for R^2 and IA when compared to the monitored concentrations for both particle size fractions, but underpredict considerably. An additional comparison of monitored against modelled concentrations by segments along the route is added in Appendix K.

6.3.4. Bus Commute

The difference between the ambient concentrations (predicting roadsides) and in-bus concentrations was adjusted by a ratio calculated from data of the mode comparison campaign (see chapter 3 and 5). The resulting concentrations for the bus commute are examined here to evaluate performance of this approach.

The focus of the result evaluation for the bus commute is on the bus route number A2/A3 (see chapter 3). Times of walking to and from bus stops, waiting at bus stop, and times in the second bus taken along the commute were excluded, as time periods in these MEs were very short (averages in these MEs are below 5min) and it is difficult to draw meaningful conclusions from such short time periods.

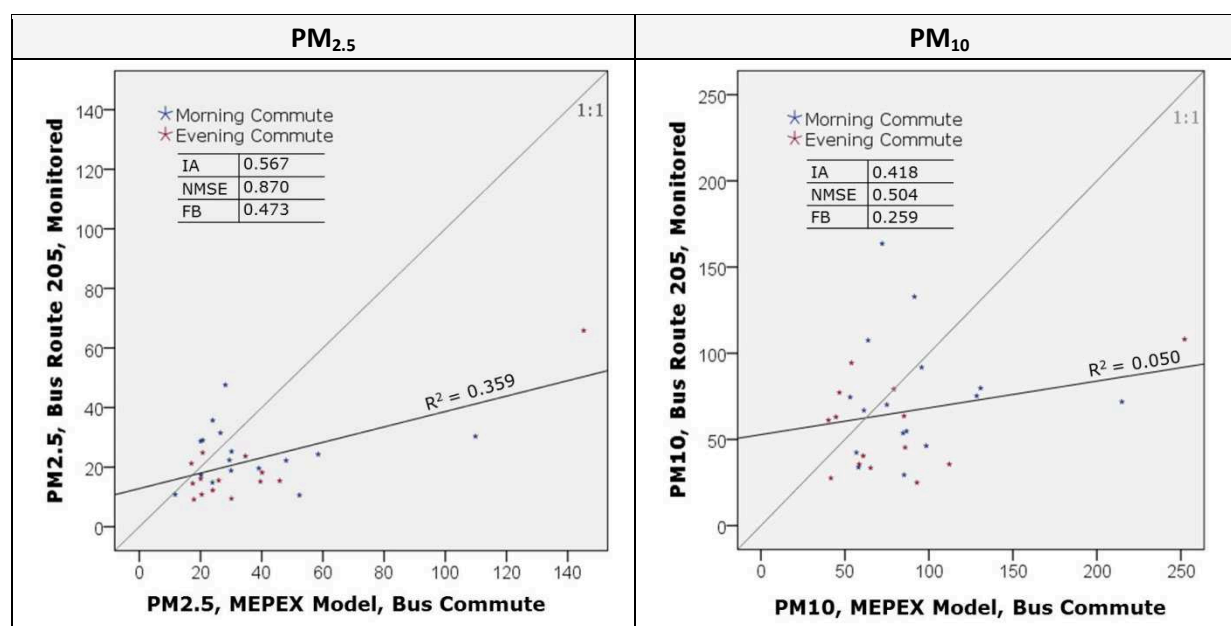


Figure 46: Modelled (MEPEX model) against Monitored PM_{2.5} and PM₁₀ concentrations (both in µg/m³) for bus commutes of individual A. 1:1 reference line and line of best fit (R^2) are added. Model performance parameters are as well presented in the graph. Both R^2 results are significant at a 0.01 level.

Model performance parameters are presented together with the scatter plots in Figure 46. For PM_{2.5} the R^2 is medium low with 0.359 and the IA shows good model fit with 0.567. The NMSE is 0.870 and the FB shows considerable overprediction with 0.473. The scatter plot however reveals that the overprediction is mainly driven by two data points.

For PM₁₀ the R^2 shows that only little of the variability could be predicted by the model with $R^2 = 0.050$. The IA in addition shows an unsatisfactory model fit of 0.418. The NMSE is with 0.504 low compared to results for other MEs; the FB indicates overprediction with 0.259. As for PM_{2.5} the scatter plot shows that the overprediction is mainly driven by two values.

The two highest predictions, both for PM_{2.5} and PM₁₀ occurred on the 18th February 2011. On this day very high background monitoring concentrations were measured (average of background stations as applied in the AVERAGE method), with 52.99 for PM_{2.5} and 62.77 for PM₁₀. The monitored personal exposure in the bus does not fully reflect the added exposure assumed by the model for

this day. A possible explanation could be that the bus wall sheltered the individual from the high particle concentrations outdoors to some extent. Other possible explanations may lie in the specific set-up inside the bus which may have caused less contribution from indoor sources compared to other journeys (e.g. fewer passengers). An additional comparison for monitored and modelled segments along the route is added in Appendix H.

6.4. Results for Personal Exposure Predictions of Individual A

In the following section results of personal exposure models are described and compared for predictions of monitored particle concentrations collected by individual A. Results are presented first for PM_{2.5}, then for PM₁₀. ME categories for visualisation of results were defined for each hour. A category was chosen for each hour according to the ME in which at least 50% of time was spent.

6.4.1. Results for PM_{2.5}

Hourly predictions

Results for model performance parameters for all models are presented in Table 57, scatter plots showing predicted against monitored personal exposure are presented in Figure 47 (Note: both axes of all plots are log scaled). Average predictions and PM_{2.5} monitoring concentrations for each hour of the day are presented in Figure 48.

For the MEPEX Model, PM_{2.5} predictions of the monitored concentrations result in a low R² of 0.113, the IA shows an unsatisfactory fit of the model with a value of 0.404, and the NMSE is high at 7.309. The FB shows overprediction with 0.197. The scatter plot for the final MEPEX model shows a generally similar trend between monitored and modelled concentrations. Scatter around the line of best fit is however high, especially for concentrations related to hours spent at home. It is evident from the scatter plot however that the two highest monitored concentrations have been extremely underpredicted by the model. The monitored concentrations in question are with 707.70µg/m³ and 327.31µg/m³ well above the predicted values of 39.19µg/m³ for the highest and 39.05µg/m³ for the second highest concentration. Both concentrations are related to cooking activities in the evening of the 18th of May 2010 (hour of cooking and hour after cooking). Data description of concentrations during and just after cooking (chapter 3) have shown that in comparison with all other cooking events for individual A this event produced the highest concentration increase and a slow return to pre-cooking concentrations. In order to test model performance without concentrations related to this particular event, performance of the dataset was evaluated without these two concentrations.

Removing the two values improved model performance noticeably and results are presented in Table 57. The model performance results without the highest two monitored concentrations show a medium low R^2 of 0.325 and a good model fit with an IA of 0.722. Removing the two values additionally lowered the NMSE to 1.629, but increased overprediction to an FB of 0.329. High concentrations hold a high risk of health effects and their correct prediction is therefore considered of particular importance. In consequence it was preferable to compare results for model performance parameters between different models including the two high values.

Approach	Method	R^2	IA	NMSE	FB
Models Based on Ambient	AVERAGE	0.020**	0.183	7.067	0.348
	NEAR	0.019**	0.208	6.494	0.469
	AMB_h	0.020**	0.145	8.794	0.103
	AMB-ta	0.018**	0.192	7.763	0.273
Modified ME Based Models	ME-h	0.055**	0.274	9.877	-0.036
	ME-o	0.084**	0.369	6.364	0.395
	MEPEX-res	0.141**	0.400	7.598	0.113
MEPEX	MEPEX	0.113**	0.404	7.309	0.197
	MEPEX (-2 outlier)	0.325**	0.722	1.629	0.329

Table 57: Model performance parameters (R^2 , IA, NMSE, and FB) for hourly estimates of $PM_{2.5}$ (in $\mu g/m^3$) monitored by individual A, using several methods for exposure prediction: AVERAGE, NEAR, AMB-h, AMB-ta, ME-h, ME-o, MEPEX-res, and MEPEX. ** $p < 0.01$

All four approaches which use ambient concentrations to predict personal exposure concentrations (AVERAGE, NEAR, AMB-h, AMB-ta) clearly show the lowest R^2 and IA results in comparison with methods including MEs (ME-h, ME-o, MEPEX-res, MEPEX). In addition the scatter plots of these models show little to no relation between monitored and predicted concentrations. All four methods using ambient concentrations to predict personal exposure show similarly low results for R^2 between 0.018 and 0.020. The IA range from 0.145 to 0.208 with the highest IA for the method using the nearest monitoring station (NEAR) for predictions. The NMSE are high for all four models with between 6.494 and 8.794, but are within the range of NMSE result of the other methods applied. The four methods all overpredict and FB results range between 0.103 for model AMB-h to 0.469 for predictions by the nearest monitoring station. The averages for each hour of the day, presented in Figure 48 show a comparably static picture throughout the day for concentrations of AVERAGE, NEAR, and AMB-h prediction methods with generally higher concentrations during daytime hours. Model AMB-ta however clearly shows peaks during times of commute and higher concentrations for the office location, compared to the home location.

Model ME-h was developed to test the influence of applying a simple I/O ratio instead of a MB model for times at home. Compared to the MEPEX model using an I/O ratio for indoor home

locations clearly reduces performance showing a much lower R^2 of 0.055 (compared to 0.113) and a reduced IA of 0.274 (compared to 0.404), the NMSE is with 9.877 the worst of all approaches. The FB is close to zero with -0.036. Looking at the average per hour in Figure 48 it is evident that the main difference to the MEPEX model occurs for the evening peak concentrations during times of cooking, which are not reflected in the ME-h model.

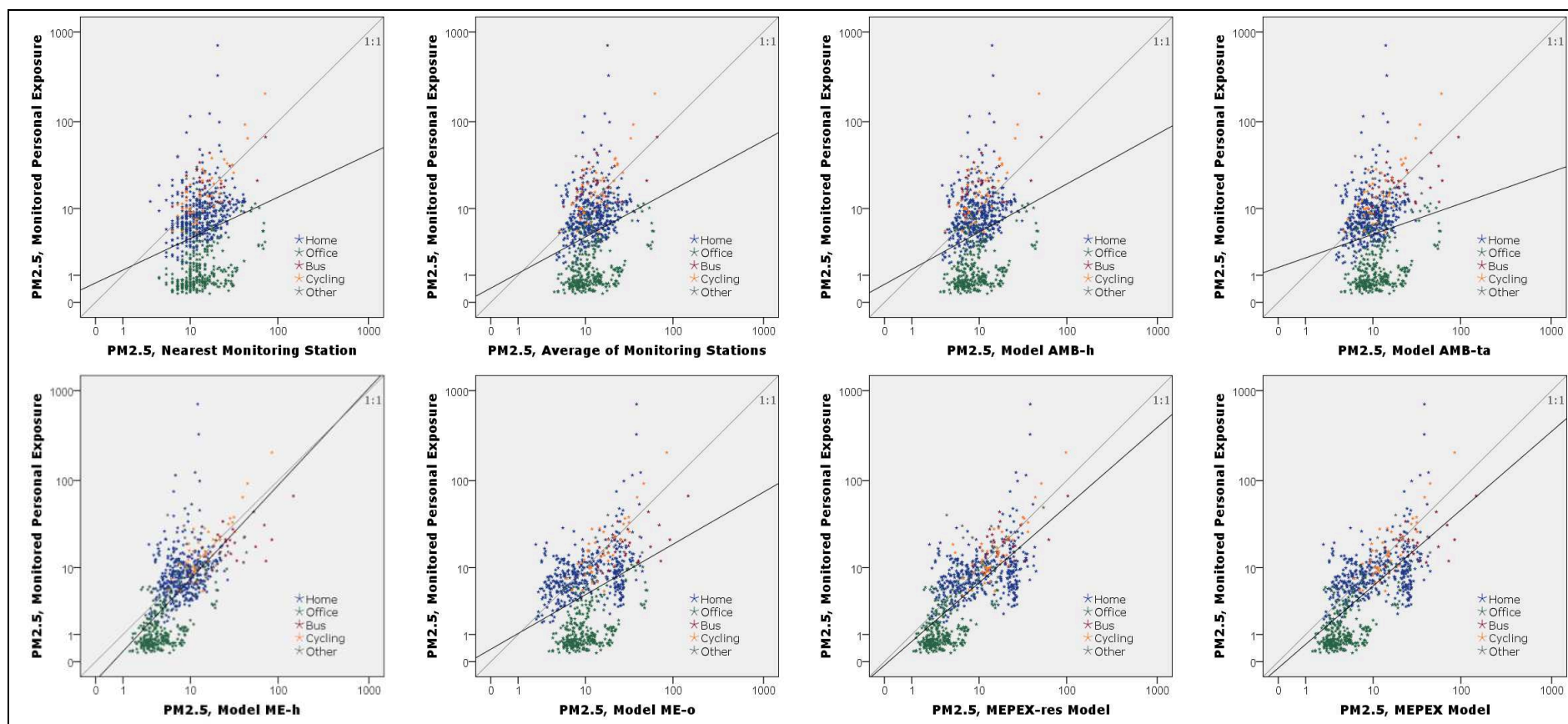


Figure 47: Predicted against monitored concentrations (individual A) for hourly $PM_{2.5}$ concentrations (in $\mu g/m^3$) for different methods of prediction: AVERAGE, NEAR, AMB-h, AMB-ta, ME-h, ME-o, MEPEX-res, and MEPEX. Both X and Y axes are presented in log scale. Reference line (1:1), as well as line of best fit are added to the graphs.

The ME-o model compares the impact of applying a home I/O ratio for the office ME as part of the personal exposure model, instead of the I/O ratio particularly calculated for work environments. Model performance parameters for R^2 and IA lie within 0.084 and 0.369 respectively below results for the MEPEX model ($R^2 = 0.113$, IA = 0.404). The NMSE (6.364) is lower than for the MEPEX model (NMSE = 7.309). The FB for the ME-o model indicates a considerable overprediction with 0.395, almost double that of the FB calculated for the MEPEX model (FB = 0.197). The averages per hour of day (Figure 48) clearly show the overprediction during times in the office compared to monitored concentrations. The overprediction of hours mostly spent in the office can additionally be clearly seen in the scatter plot (Figure 47). In summary, using the home I/O ratio for times in the office increased already too high predictions for the office ME, which in turn lowered overall model performance for R^2 and IA.

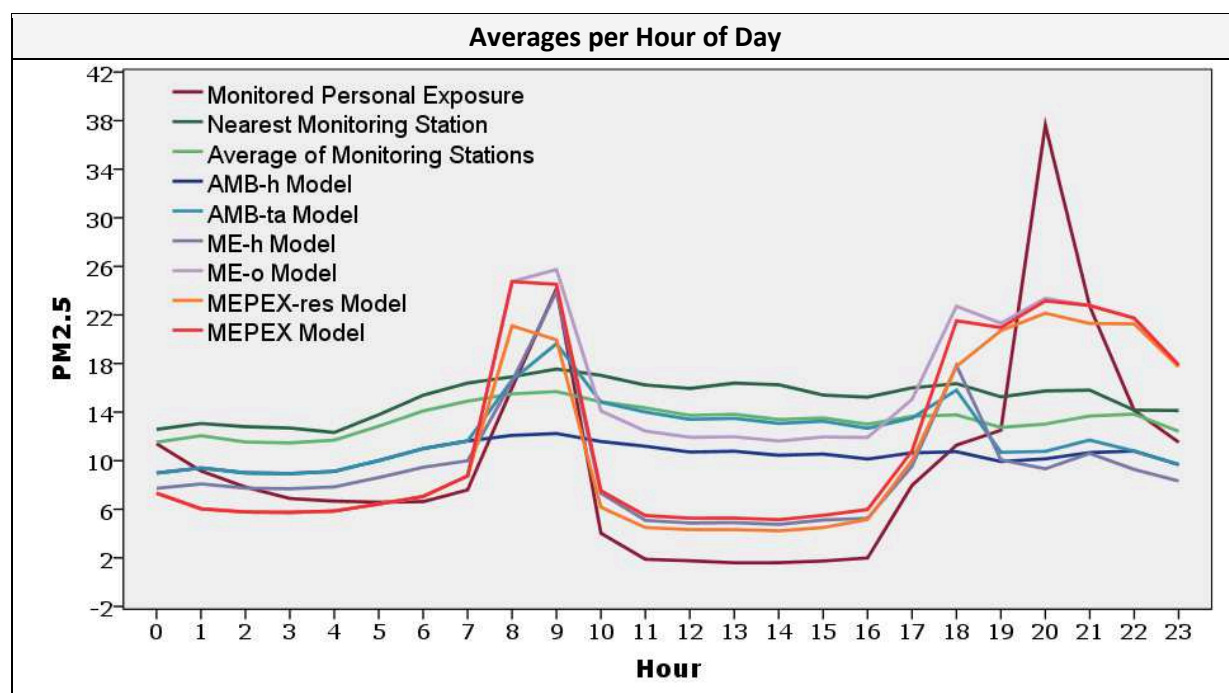


Figure 48: PM_{2.5} concentrations (µg/m³) by hour of day for monitored personal exposure of individual A and results for different prediction methods of personal exposure (AVERAGE, NEAR, AMB-h, AMB-ta, ME-h, ME-o, MEPEX-res, and MEPEX).

Model performance parameter, as well as the scatter plot of the MEPEX-res model show similar results compared to the MEPEX model. R^2 results of the MEPEX-res model are a little higher (0.141) compared to the MEPEX model ($R^2 = 0.113$). The IA (0.400) is almost the same as the IA of the MEPEX model with 0.404. The NMSE of the MEPEX-res model is slightly higher (7.598) compared to the MEPEX model (NMSE = 7.309) and the FB shows a less pronounced overprediction for the

MEPEX-res model with 0.113 (compared to 0.197). Results per hour of day shows as well little difference between MEPEX-res and MEPEX model, slightly lower concentrations can however be observed for the MEPEX-res model during times in commute and in the office.

Predictions of Time-Weighted Daily Exposure:

The model performance parameters of daily predictions for personal exposure of individual A are presented in Table 58 for all applied methods. Scatter plots of modelled against monitored are presented in Figure 49. Generally results show that model performance for daily estimates shows better model performance compared to hourly estimates.

The MEPEX model shows generally good performance for daily exposure predictions with an R^2 of 0.439, an IA of 0.661 indicating good model fit. Results also show the lowest NMSE compared with the other methods of 0.608. The FB shows an overprediction of 0.208. The scatter plots of all methods show one very high monitored concentration of $60.38\mu\text{g}/\text{m}^3$. This high daily average was monitored on the 18th of May 2010. The daily average concentration for that day at London background monitoring sites (average of sites as applied in AVERAGE method) was elevated with $22.04\mu\text{g}/\text{m}^3$ compared to the mean concentration for all hours of this analysis of $13.49\mu\text{g}/\text{m}^3$. In addition, cooking during the 18th of May 2010 resulted in the highest monitored hourly concentrations for individual A (see above). The combination of both elevated background concentration and high exposure during cooking resulted in a daily personal exposure well above average, for which all methods showed limited ability to predict. Removing the high value from the dataset was investigated, but does not improve results for model performance parameters.

Model performance parameters for daily exposure clearly show the lowest R^2 for the four methods using only ambient concentrations for predictions (between 0.117 and 0.173). The scatter plots of these methods all show little agreement between monitored and predicted. The IA of these methods is for AMB-h and AMB-ta also the lowest of all prediction methods with 0.471 and 0.443 respectively. Results for IA of the AVERAGE and NEAR methods lie above results for the ME-h model, but below results of all other models (ME-o, MEPEX-res, and MEPEX). The IA for the NEAR method lies at just below 0.5 with 0.496, the IA for the AVERAGE method lies just above 0.5 with 0.511, indicating good model fit. The NMSE of the models using ambient concentrations for personal exposure predictions show a comparably low error between 0.787 and 0.860. The estimates show overprediction with FB between 0.110 and 0.472 with the highest overprediction by the NEAR method. No improvement could be observed between the AMB-h and the AMB-ta method for adding time-activity locations.

Approach	Method	R ²	IA	NMSE	FB
Models Based on Ambient	AVERAGE	0.173**	0.511	0.787	0.355
	NEAR	0.149**	0.496	0.860	0.472
	AMB_h	0.173**	0.471	0.853	0.110
	AMB-ta	0.117**	0.443	0.846	0.253
Modified ME Based Models	ME-h	0.223**	0.487	0.970	-0.071
	ME-o	0.303**	0.592	0.696	0.393
	MEPEX-res	0.489**	0.687	0.614	0.177
MEPEX		0.439**	0.661	0.608	0.208

Table 58: Model performance parameters (R², IA, NMSE, and FB) for daily estimates of PM_{2.5} (µg/m³) monitored by individual A, using several methods for exposure prediction: AVERAGE, NEAR, AMB-h, AMB-ta, ME-h, ME-o, MEPEX-res, and MEPEX. ** p < 0.01

The ME-h model shows, as for hourly concentrations, reduced performance results compared to the MEPEX model. The use of an I/O ratio instead of using the MB model clearly had a large impact reducing the R² to 0.223 (compared to 0.439) and showing a model fit below 0.5 of 0.487 compared to 0.661 for the MEPEX model. The NMSE is with 0.970 the highest for all methods applied. The FB indicates only slight underprediction with -0.071. Changing the office I/O ratio to a home I/O ratio for model ME-o results reduced performance parameters compared to the MEPEX model, showing an R² of 0.303, an IA of 0.592, and an NMSE of 0.696. In addition, the model showed higher overprediction, which is reflected by an FB of 0.393.

The MEPEX-res model shows model performance results similar to, and for some parameters slightly above, results for the MEPEX model. The R² result and the result for the IA lie slightly above results for the MEPEX model with 0.489 and 0.687, respectively. The NMSE is with 0.614 slightly higher compared to the MEPEX model. The FB shows in comparison less pronounced overprediction with 0.177. Scatter plots of the two modelling approaches (MEPEX and MEPEX-res) also show very similar results.

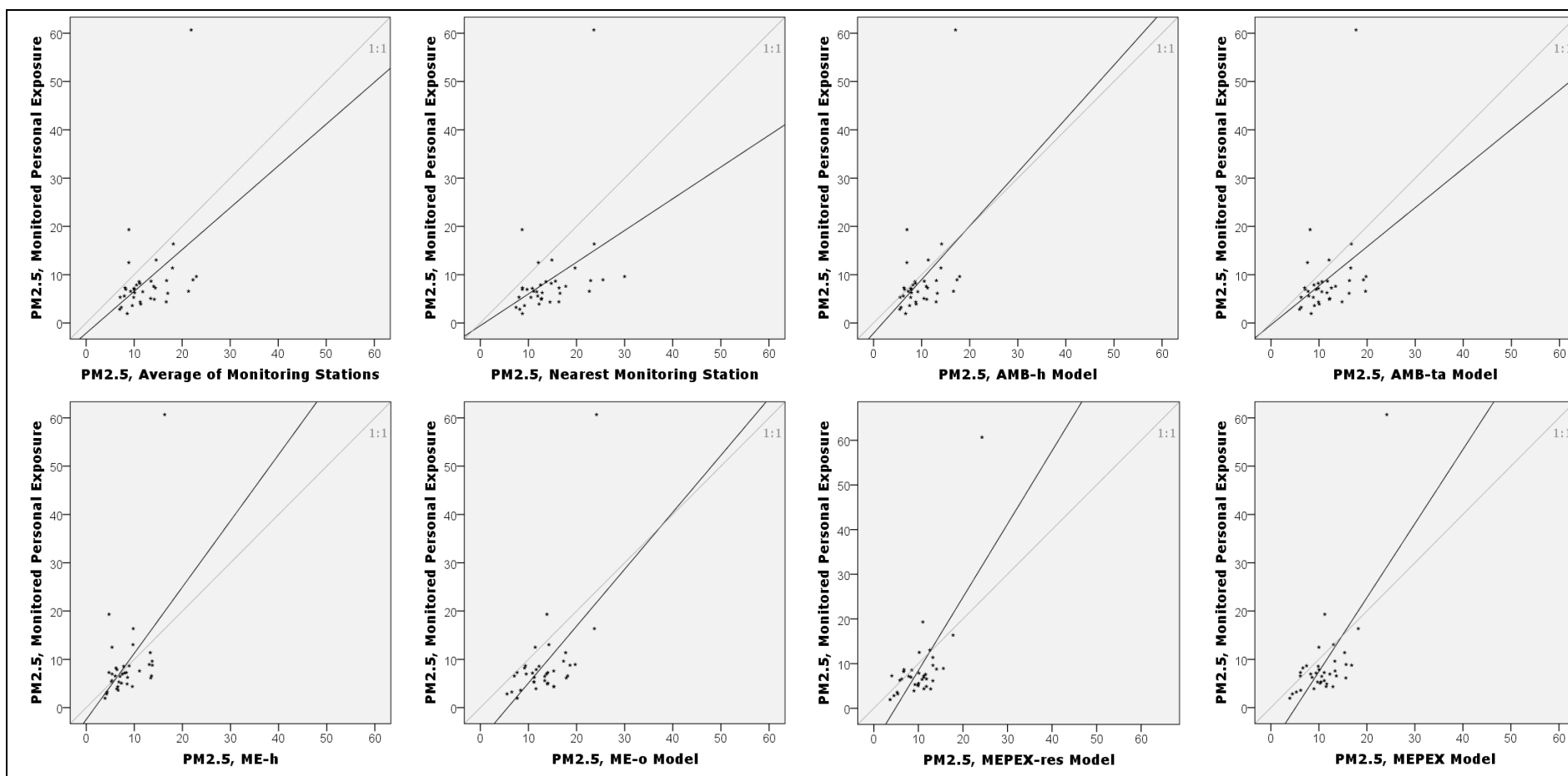


Figure 49: Predicted against monitored concentrations (individual A) of daily PM_{2.5} concentrations (in $\mu\text{g}/\text{m}^3$) for different methods of prediction: AVERAGE, NEAR, AMB-h, AMB-ta, ME-h, ME-o, MEPEX-res, and MEPEX. Reference line (1:1), as well as line of best fit are added to the graphs.

The seasonal averages for all methods of prediction and the monitored personal exposure are presented in Figure 50. The graph shows that for all monitored and predicted concentrations the lowest seasonal concentrations occurred during summer. All models show the second lowest concentrations during autumn. Most prediction approaches show the highest concentrations during winter and spring with little difference between these two seasons. The nearest monitoring stations however showed clearly lower concentrations in spring compared to winter. For the monitored personal exposure the second lowest seasonal average occurs in winter, followed by autumn and the highest seasonal average is in spring. It should be noted that one of the eight days in spring is the 18th of May, which showed much higher concentrations for monitored personal exposure than any other day. This day most likely increased average PM_{2.5} concentrations of personal exposure monitoring for spring.

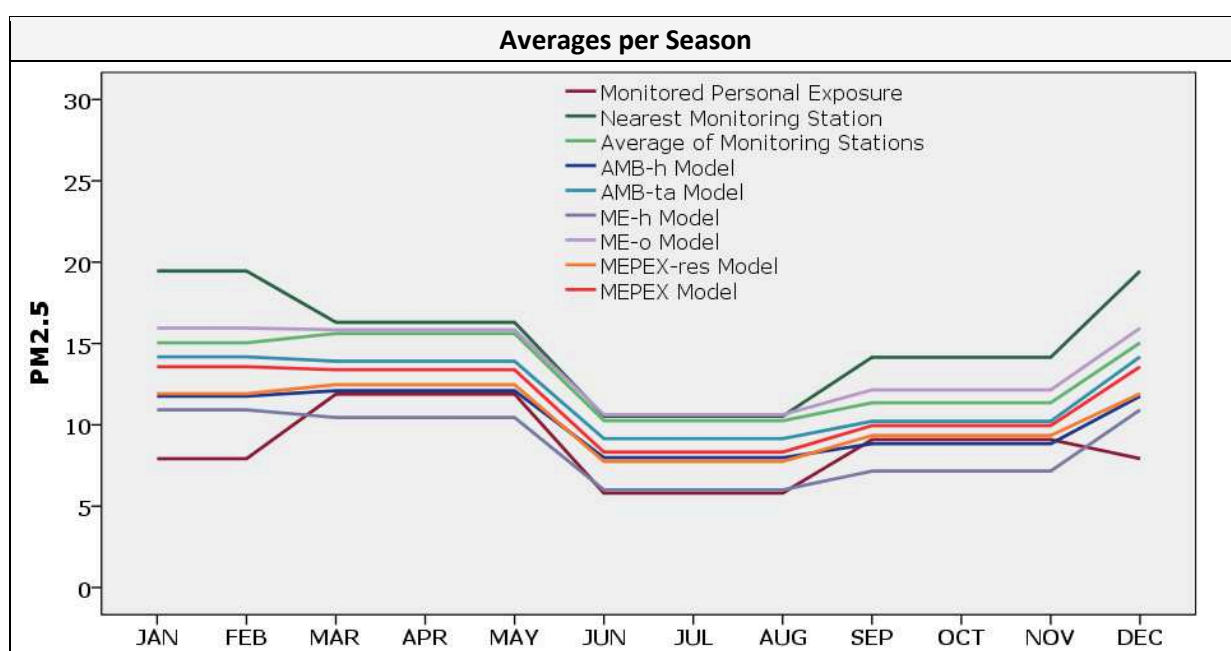


Figure 50: PM_{2.5} concentrations (µg/m³) by season for monitored personal exposure of personal exposure of individual A and results for different prediction methods (AVERAGE, NEAR, AMB-h, AMB-ta, ME-h, ME-o, MEPEX-res, and MEPEX).

6.4.2. Results for PM₁₀

Hourly Predictions

For hourly PM₁₀ predictions of personal exposure for concentrations collected for individual A, model performance parameters are presented in Table 59, scatter plots of predicted against monitored concentrations are presented in Figure 52 and average concentrations per hour of day are presented in Figure 51.

Model performance parameters of the MEPEX model show good model performance with an IA result of 0.629. The R^2 is however low with 0.210 and the error is high with an NMSE of 5.948. The FB result indicates considerable overprediction with 0.396.

Approach	Method	R^2	IA	NMSE	FB
Models Based on Ambient	AVERAGE	0.006*	0.136	9.335	-0.103
	NEAR	0.001*	0.097	7.853	0.114
	AMB_h	0.006**	0.086	11.086	-0.270
	AMB-ta	0.013**	0.110	10.377	-0.214
Modified ME Based Models	ME-h	0.040**	0.195	11.382	-0.327
	ME-o	0.202**	0.617	5.455	0.498
	ME-t	0.200**	0.621	6.284	0.342
	MEPEX-res	0.182**	0.598	6.167	0.470
MEPEX		0.210**	0.629	5.948	0.396

Table 59: Model performance parameters (R^2 , IA, NMSE, FB) for hourly estimates of PM_{10} (in $\mu g/m^3$) monitored by individual A, using several methods for exposure prediction: AVERAGE, NEAR, AMB-h, AMB-ta, ME-h, ME-o, MEPEX-res, and MEPEX. * significant at a 0.05 level, ** $p < 0.01$

The four methods using predictions based only on ambient concentrations generally show much lower results for model performance parameters in comparison to approaches including ME models. The scatter plots of these four methods show a similar data pattern without clear relationship between the monitored and predicted concentrations. R^2 results for the methods are very low and range between 0.001 and 0.013, meaning that close to none of the variability in the data could be explained by the methods. Results for the IA also show a low model fit with between 0.086 and 0.136 of these methods. NMSE results are very high with between 7.853 and 11.086. The FB shows underprediction for the AVERAGE, AMB-h, and AMB-ta methods with -0.103, -0.270, and -0.214 respectively. Using the NEAR method however resulted in overprediction with an FB of 0.114. Underlying the overprediction of the NEAR method is a generally higher average for the concentrations at the nearest monitoring site (NEAR), compared to the average of monitoring sites (AVERAGE), which can be observed in Figure 51. The graph also clearly shows the difference between the AMB-h and AMB-ta model, with average hourly concentrations of the AMB-ta model increased during times of commute. Between the AMB-h model and the AMB-ta model a small increase of R^2 and IA, as well as reduction of the NMSE and underprediction can be observed. This indicates that adding location to the model causes a small improvement in performance.

The application of the ME-h model tests the difference of using an I/O ratio to model indoors at home instead of a mass-balance (MB) model. Predictions of the ME-h model result in a very low R^2 of 0.040, and an IA of 0.195, which is well below a good model fit ($IA > 0.5$). In addition the NMSE shows the highest error compared to all other methods with 11.382. The change of the indoor

model evidently caused a clear reduction in model performance compared to the MEPEX model (MEPEX model: $R^2 = 0.210$, IA = 0.629, NMSE = 5.948). The FB shows underprediction with an FB of -0.327. This shows that the overprediction observed for the MEPEX model is to a great extent related to the application of the MB model to predict home indoor concentrations. The scatter plot of the ME-h model clearly shows a linear relationship between monitored and modelled concentrations. Underprediction of several high concentrations for the home indoor ME can also be observed. The averages for each hour of the day show most clearly that the lack of a situation resolved indoor model (e.g. including cooking, active occupancy) causes large portions of the daily variability of personal exposure to be greatly underpredicted.

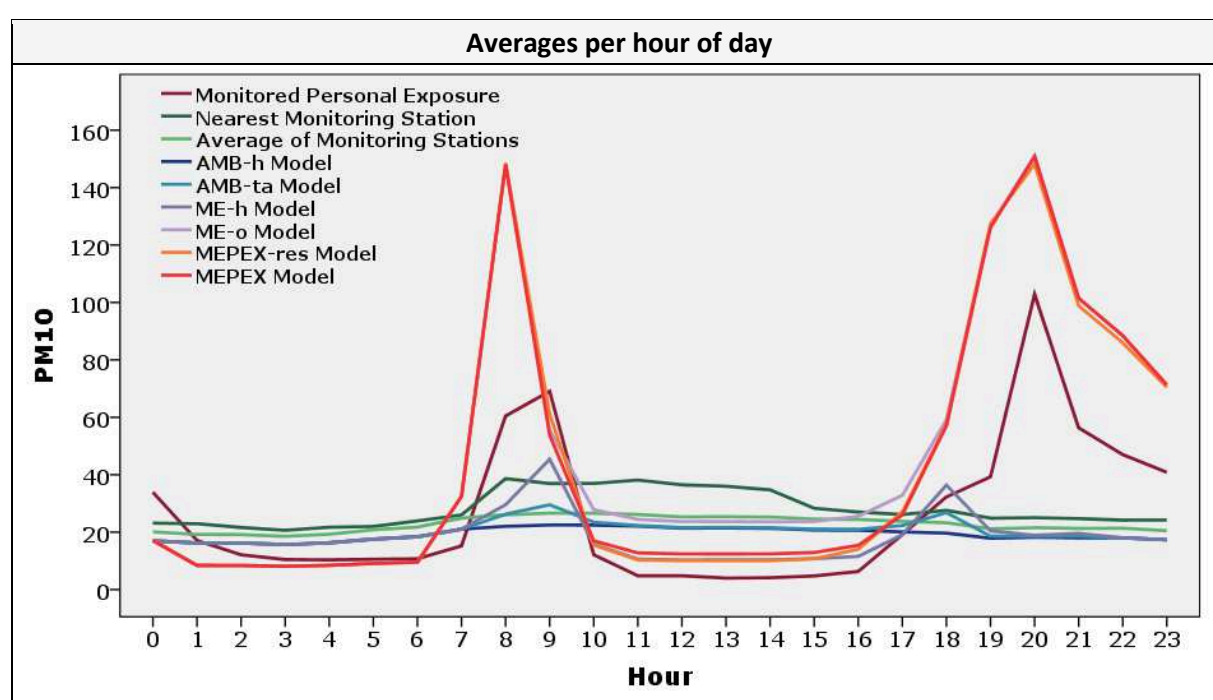


Figure 51: PM₁₀ concentrations (µg/m³) by hour of day for monitored personal exposure of personal exposure of individual A and results for different prediction methods (AVERAGE, NEAR, AMB-h, AMB-ta, ME-h, ME-o, MEPEX-res, and MEPEX).

The ME-h model tested the effect of using an I/O ratio for indoor homes instead of the office I/O applied in the MEPEX model. The home I/O ratio is higher than the office I/O ratio, which already resulted in overprediction for the office ME. The effect of this additional overprediction of times in the office can clearly be seen in the scatter plot and causes a raised FB to 0.498. The effect on the general model performance is however only a reduction of the R^2 by 0.008 to 0.202 compared to the MEPEX model and a reduction by 0.008 to 0.617 for the IA result. The NMSE is with 5.455 slightly below the result of the MEPEX model (NMSE = 5.948).

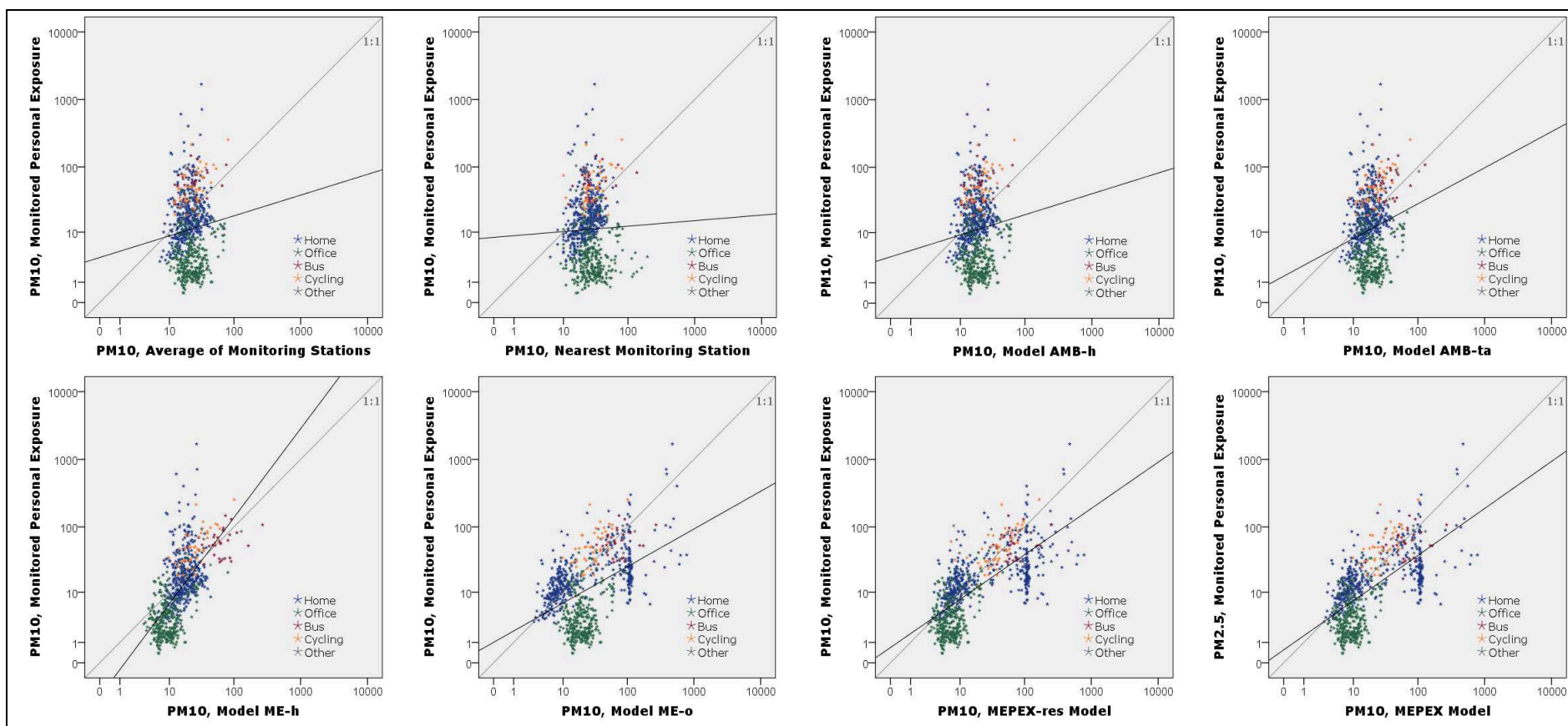


Figure 52: Predicted against monitored concentrations (individual A) for hourly PM₁₀ concentrations (in $\mu\text{g}/\text{m}^3$) for different methods of prediction: AVERAGE, NEAR, AMB-h, AMB-ta, ME-h, ME-o, MEPEX-res, and MEPEX. Both X and Y axes are presented in log scale. Reference line (1:1), as well as line of best fit are added to the graphs.

The MEPEX-res model shows model performance results slightly below the MEPEX model with a low R^2 of 0.182, an IA of 0.598, indicating good model fit. The NMSE is also in comparison a little higher with 6.167. The MEPEX-res model additionally shows a higher overprediction with an FB of 0.470. Little difference between the MEPEX-res model and the MEPEX models can be observed from the scatter plots or between differences between hours of day.

Predictions of Time-Weighted Daily Exposure:

Model performance parameters for daily averages of PM_{10} have been summarised in Table 60. Scatter plots of predicted against monitored concentrations are presented in Figure 53. As for $PM_{2.5}$, the daily predictions generally show better results for model performance compared to hourly predictions. The MEPEX model shows an R^2 at a medium level of 0.403 and a good model fit of 0.725. The error is comparably low with an NMSE of 0.657. The model shows considerable overprediction with an FB of 0.451. The scatter plot clearly shows a linear relationship between the MEPEX model predictions and monitored concentrations.

It also shows that the highest monitored concentration was considerably underpredicted. This concentration (of $149.74\mu\text{g}/\text{m}^3$) was measured on the 18th of May 2010. As for the $PM_{2.5}$ results, high concentrations on this day are most likely related to 1) particularly high background concentrations for PM_{10} at all London background concentrations with $34.32\mu\text{g}/\text{m}^3$ compared to $23.12\mu\text{g}/\text{m}^3$ for the average of all hours used in this application (average at monitoring stations, as selected for the AVERAGE method), and 2) high concentrations during cooking.

Approach	Method	R^2	IA	NMSE	FB
Models Based on Ambient	AVERAGE	0.086**	0.362	0.990	-0.106
	NEAR	0.020**	0.287	0.880	0.124
	AMB_h	0.086**	0.282	1.241	-0.273
	AMB-ta	0.084**	0.285	1.172	-0.231
Modified ME Based Models	ME-h	0.145**	0.345	1.369	-0.367
	ME-o	0.374**	0.679	0.745	0.543
	MEPEX-res	0.409**	0.708	0.684	0.514
MEPEX		0.403**	0.725	0.657	0.451

Table 60: Model performance parameters (R^2 , IA, NMSE, FB) for daily estimates of PM_{10} ($\mu\text{g}/\text{m}^3$) monitored by individual A, using several methods for exposure prediction: AVERAGE, NEAR, AMB-h, AMB-ta, ME-h, ME-o, MEPEX-res, and MEPEX. ** $p < 0.01$

In comparison with the MEPEX model, methods using any form of prediction based only on ambient concentrations showed weak model performance. R^2 for the AVERAGE, NEAR, AMB-h, and AMB-ta methods range between 0.020 and 0.086 and all IA results are with between 0.282 and 0.362 well

below a good model fit (indicated by an IA of above 0.5). Results for NMSE lie between 0.880 and 1.172. As for hourly concentrations, the AVERAGE, AMB-h, and AMB-ta models underpredict and FB lie between -0.106 and -0.273, the FB of the NEAR model shows overprediction with 0.124. Little difference between results for model parameters can be observed between AMB-h (only home) and the AMB-ta model (with added spatial location).

Results for model performance parameters of the ME-h model are weaker than results for the MEPEX model. The R^2 is low with 0.145 (compared to 0.403) and the IA is with 0.345 even lower than for the ambient-based AVERAGE method. The NMSE is the highest observed for any of the methods with 1.369 and the FB shows a considerable underprediction with -0.367.

Model performance results for the ME-o model show a small decrease in performance compared to the MEPEX model. Both R^2 and IA are slightly lower compared to the MEPEX model with 0.374 and 0.679 respectively. NMSE and FB results are compared with the MEPEX model a little higher with 0.745 and 0.543 respectively.

Model performance results from the MEPEX-res model are similar to results from the MEPEX model with a medium R^2 of 0.409, and an IA of 0.708, which indicates a good model fit. NMSE, as well as FB are slightly higher compared to the MEPEX model with 0.684 and 0.514 respectively. Little difference between MEPEX-res and MEPEX model predictions can be observed from the scatter plots.

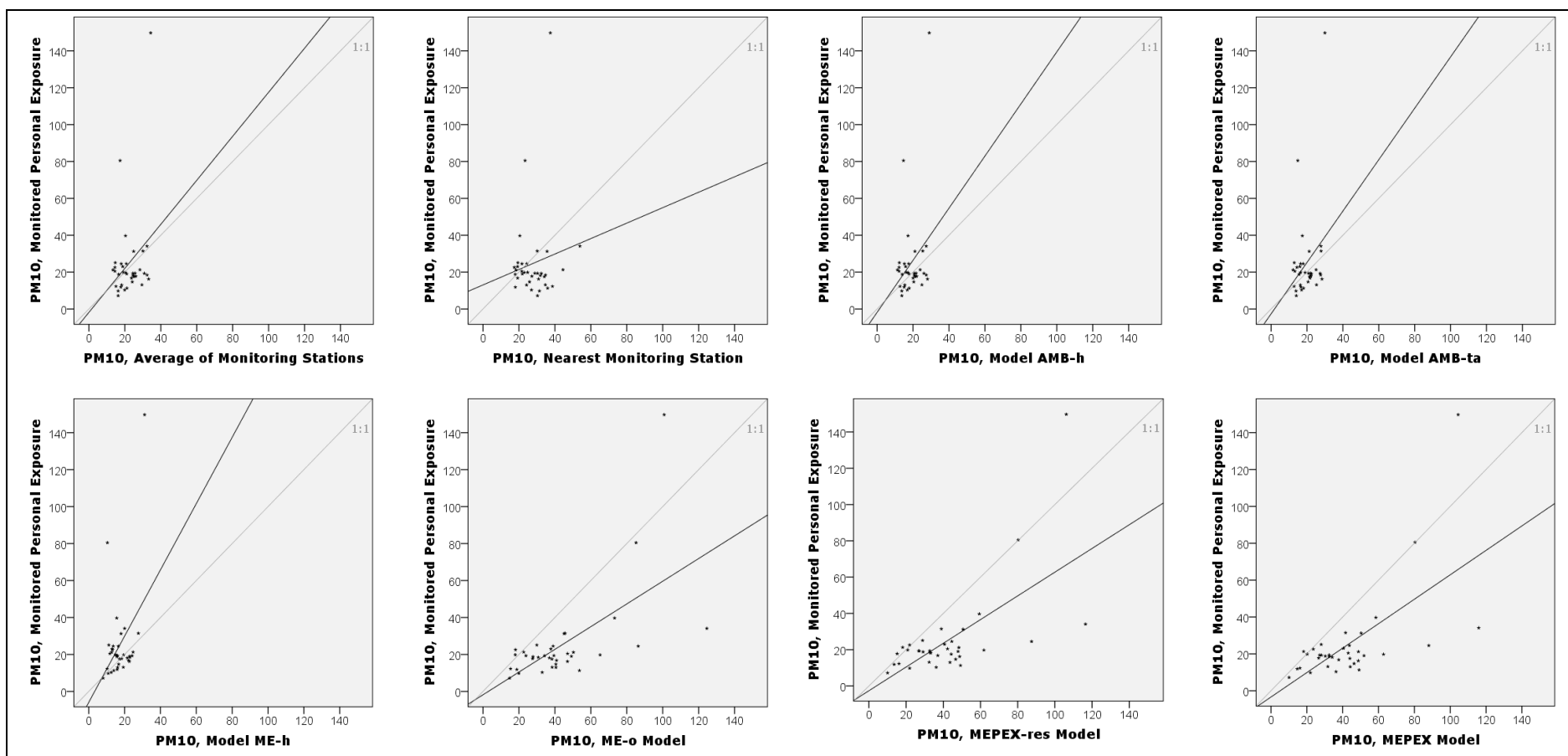


Figure 53: Predicted against monitored concentrations (individual A) for daily PM₁₀ concentrations (in $\mu\text{g}/\text{m}^3$) for different methods of prediction: AVERAGE, NEAR, AMB-h, AMB-ta, ME-h, ME-o, MEPEX-res, and MEPEX. Both X and Y axes are presented in log scale. Reference line (1:1), as well as line of best fit are added to the graphs.

Seasonal differences, as reflected for the days of personal monitoring collected by individual A are presented in Figure 54. Most prediction methods, as well as the monitored exposure show the lowest seasonal average in summer. For the AVERAGE, the AMB-h, the AMB-ta, and the ME-h method concentrations are a little higher for the other three seasons. The ME-o, MEPEX-res and MEPEX model show highest concentration for the winter average. The average concentrations for these three models are generally much higher than for any of the other prediction methods or the monitored concentrations. The nearest monitoring station is the only prediction method, which does not show the lowest seasonal average for summer. Concentrations at the nearest station show lowest concentrations in winter and highest concentrations in autumn. The monitored personal exposure shows, as for PM_{2.5} highest concentrations in spring. As before, it is likely that the raised concentrations on the 18th of May 2010 increased the monitored spring average concentration.

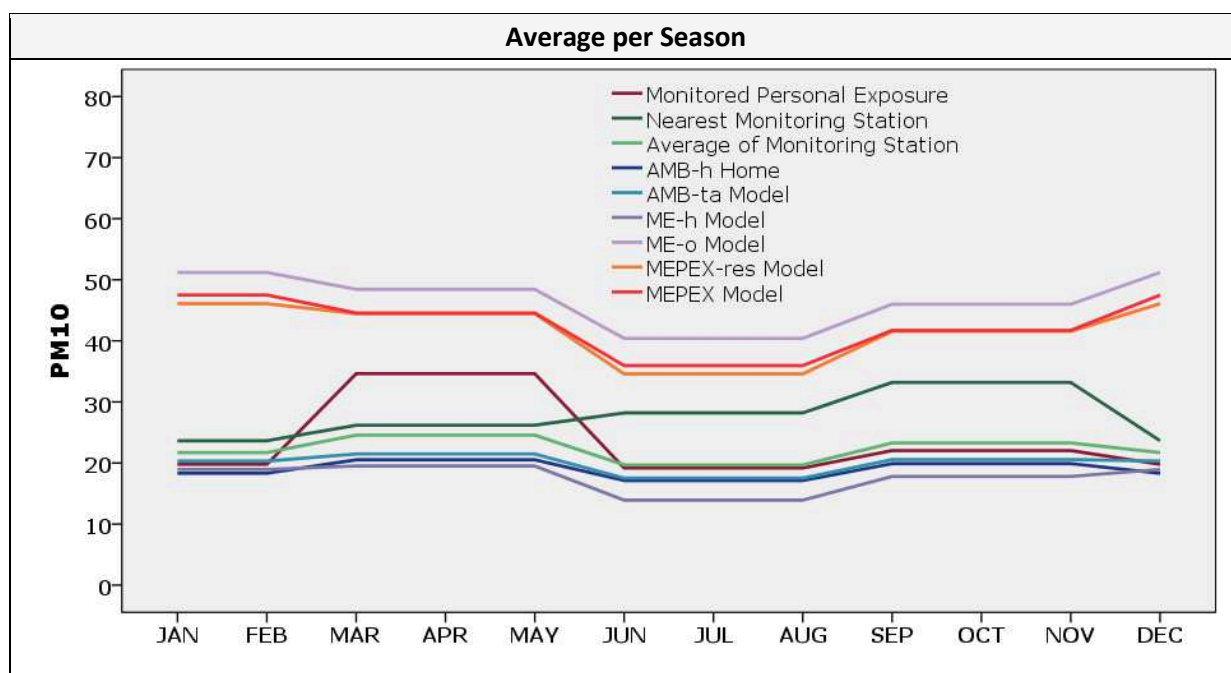


Figure 54: PM₁₀ concentrations (µg/m³) by season for monitored personal exposure of individual A and results for different prediction methods of personal exposure (AVERAGE, NEAR, AMB-h, AMB-ta, ME-h, ME-o, MEPEX-res, and MEPEX).

6.4.3. Result Summary

The results presented above for the prediction of personal exposure for individual A clearly show that for both particle size fractions the MEPEX model and the MEPEX-res model have predicted personal exposure best, compared to other approaches. The comparison to other approaches suggests that the MEPEX model may be a useful tool for future applications to predict individual

exposure. The MEPEX-res model may in addition be considered as a simpler alternative for exposure modelling. To test transferability of both MEPEX and MEPEX-res models for exposure predictions of other individuals (apart from individual A), these two models have therefore been selected to predict personal exposure monitored for individuals B, C, and D (introduced in chapter 3) in the next section. In order to show model applicability for a larger population, this is followed by an application of the MEPEX-res model to predict personal exposure for a study population in London. All results are discussed at the end of this chapter.

6.5. Results for Personal Exposure Predictions of Individuals B, C, and D

MEPEX and MEPEX-res model have been applied to predict hourly personal exposure of individuals B, C, and D. Only small datasets were collected for each of these individuals during two days for individuals B, and C, and during four days for individual D. The dataset for individual C only includes 16 hours which were exclusively recorded during times away from home (commute by walking and bus, as well as time in the office and at a nursery). The data of individual C was therefore not considered fully representative of daily exposure. In consequence, data for individual C is included in the scatter plots, but model performance results of IA, NMSE, and FB were not included.

Scatter plots for predictions of the MEPEX and MEPEX-res models for hourly monitored personal concentrations of individuals B, C, and D are presented in Figure 55; model performance parameters for individuals B and D are added. For the MEPEX model, results for individual B (47 hours) show generally weak performance with an R^2 of 0.083 for $PM_{2.5}$ and an R^2 of 0.052 for PM_{10} . IA results are also low with 0.149 for $PM_{2.5}$ and 0.064 for PM_{10} . The error is high with an NMSE of 7.738 for $PM_{2.5}$ and an NMSE of 21.690 for PM_{10} . The FB for both particle size fractions shows overprediction well above a factor of 2 with 1.160 for $PM_{2.5}$ and 1.531 for PM_{10} . The scatter plots (Figure 55) shows highest overprediction for a group of data points estimated for individual B (right hand side of the graph) both for $PM_{2.5}$ and PM_{10} . It is likely that this group of highly overpredicted data points generally lowered performance for predictions of personal exposure for individual B.

Scatter plots of the MEPEX model for individual C show a very good relationship between modelled and monitored, which is confirmed by a high R^2 of 0.881 for $PM_{2.5}$ and 0.933 for PM_{10} . Results suggest that the MEPEX model predicts better if only times away from home are considered. This may be related to the generally weak model performance observed for predictions of the applied indoor model (see chapter 5).

Model performance results of the MEPEX model for individual D (94 hours) show medium low R^2 results of 0.291 for $PM_{2.5}$ and 0.389 for PM_{10} . The results for the IA indicate good model fit with 0.697 for $PM_{2.5}$ and 0.537 for PM_{10} . The NMSE is with 1.527 much lower for $PM_{2.5}$ than for PM_{10} with 7.376. The FB indicates a comparably moderate overprediction for $PM_{2.5}$ with 0.114 and a high overprediction by more than a factor of 2 for PM_{10} with $FB = 0.923$.

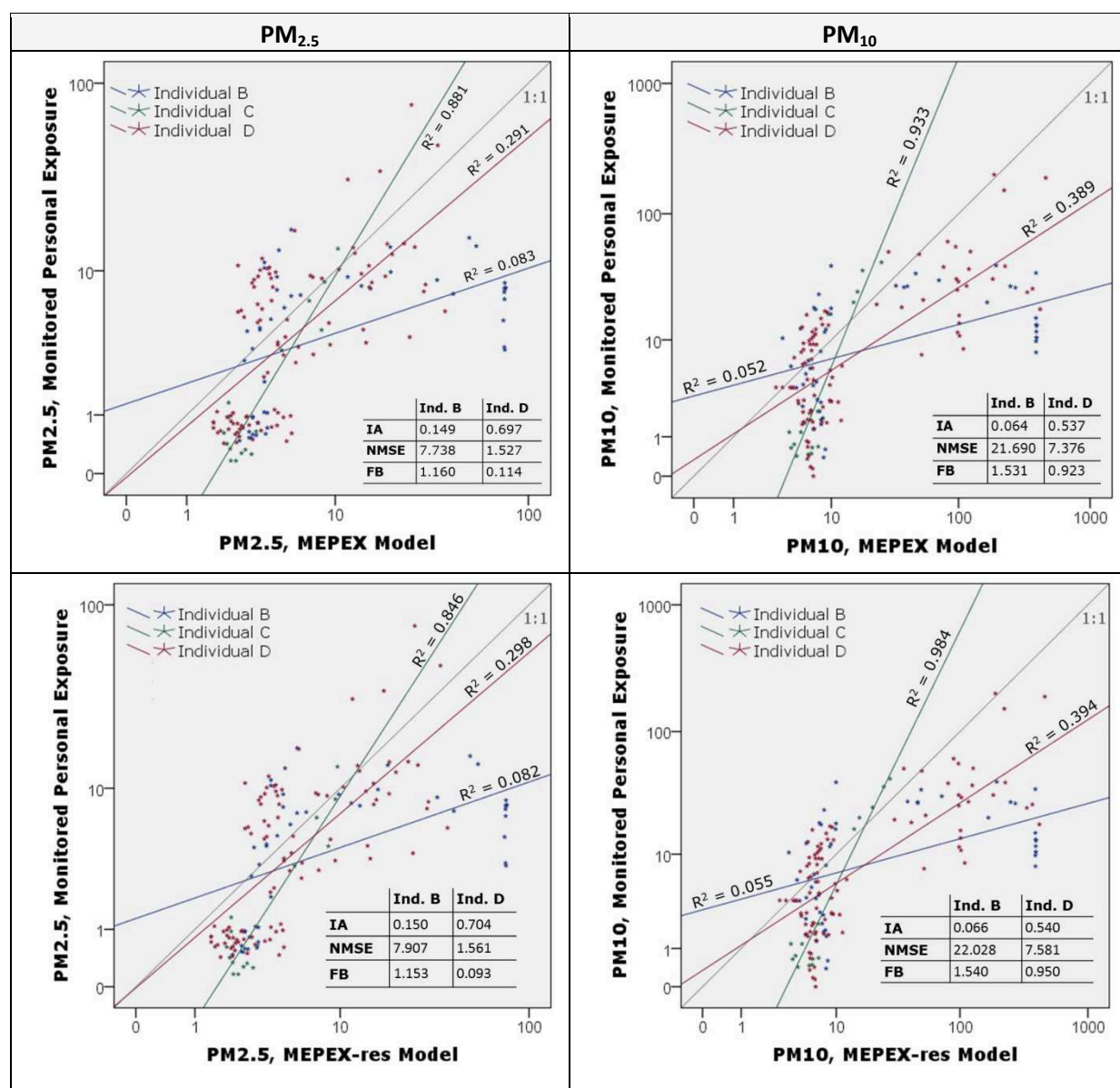


Figure 55: Modelled against monitored $PM_{2.5}$ and PM_{10} concentrations (both in $\mu g/m^3$) for hourly concentrations of personal exposure collected by individuals B, C, and D, specified by individual, for the MEPEX model (top) and the MEPEX-res model (bottom). Both X and Y axes are presented in log scale. Reference line and line of best fit per individual are added to the graph. Model performance parameters for individuals A and D are presented within the graphs.

Results for the MEPEX-res model show generally very similar results for model performance parameters compared to the MEPEX model. For individual B, results for model performance parameters are again weak both for PM_{2.5} and PM₁₀. The results for R² are 0.082 for PM_{2.5} and 0.055 for PM₁₀, which differs by less than 0.01 compared to results from the MEPEX model. Results for the IA are 0.150 for PM_{2.5} and 0.066 for PM₁₀, which again only differ marginally from results by the MEPEX model. The NMSE shows very high results of 7.907 for PM_{2.5} and 22.028 for PM₁₀, which are both slightly above results for the MEPEX model. FB results show overprediction for both size fraction with 1.153 for PM_{2.5} and 1.540 for PM₁₀, which is slightly higher than for the MEPEX model for PM_{2.5} and slightly lower than the MEPEX model for PM₁₀.

For individual C, R² results of the MEPEX-res model are 0.846 for PM_{2.5} (a little lower compared to the MEPEX model) and 0.984 for PM₁₀ (a little higher compared to the MEPEX model). For individual D, both model performance results for R² and IA for both size fraction differ by less than 0.01 from results of the MEPEX model (PM_{2.5}: R² = 0.298, IA = 0.704; PM₁₀: R² = 0.394, IA = 0.540). As for individual B, NMSE results for the MEPEX-res model of individual D are a little higher than for the MEPEX model (PM_{2.5}: NMSE = 1.561; PM₁₀: NMSE = 7.581). FB results for individual D show overprediction for both particle size fractions. For PM_{2.5} the FB is 0.093, which is slightly below results for the MEPEX model. For PM₁₀ the FB is 0.950, which is slightly above results for the MEPEX model.

The hourly predictions per ME are presented in Figure 56 (including hours predicted for individual C). Very little difference can be observed between the scatter plots of the two different models, both for PM_{2.5} and PM₁₀. In the following paragraph, the scatter plots of both MEPEX and MEPEX-res models are therefore described here together. For the office ME overprediction both for PM_{2.5} and for PM₁₀ can be observed. The few hours spent predominately in commute (bus, walking) show predictions close to the reference line. Times spent in the home indoor ME are for some data points extremely overpredicted, especially for PM₁₀. The group of overpredicted data points identified for individual B, above, falls into the indoor category. Individual B did not cook during the personal exposure monitoring campaign and investigation of the relevant dates did not show increased ambient concentrations. The overprediction therefore most likely occurred by overpredicting the effect of active occupancy for the indoor ME. The MB model uses home volume for the estimation of source contributions to the indoor environment. Home B is with 40.50m³ very small. A smaller home volume increases source contributions for modelled concentrations (less particle dispersion), which may have resulted in overpredictions.

The similarity between results of the MEPEX model and the MEPEX-res model had been observed for individual A. The observation of similar results for both models when predicting for other individuals

confirms that the MEPEX-res model may be a simpler, but equally well performing alternative to the MEPEX model.

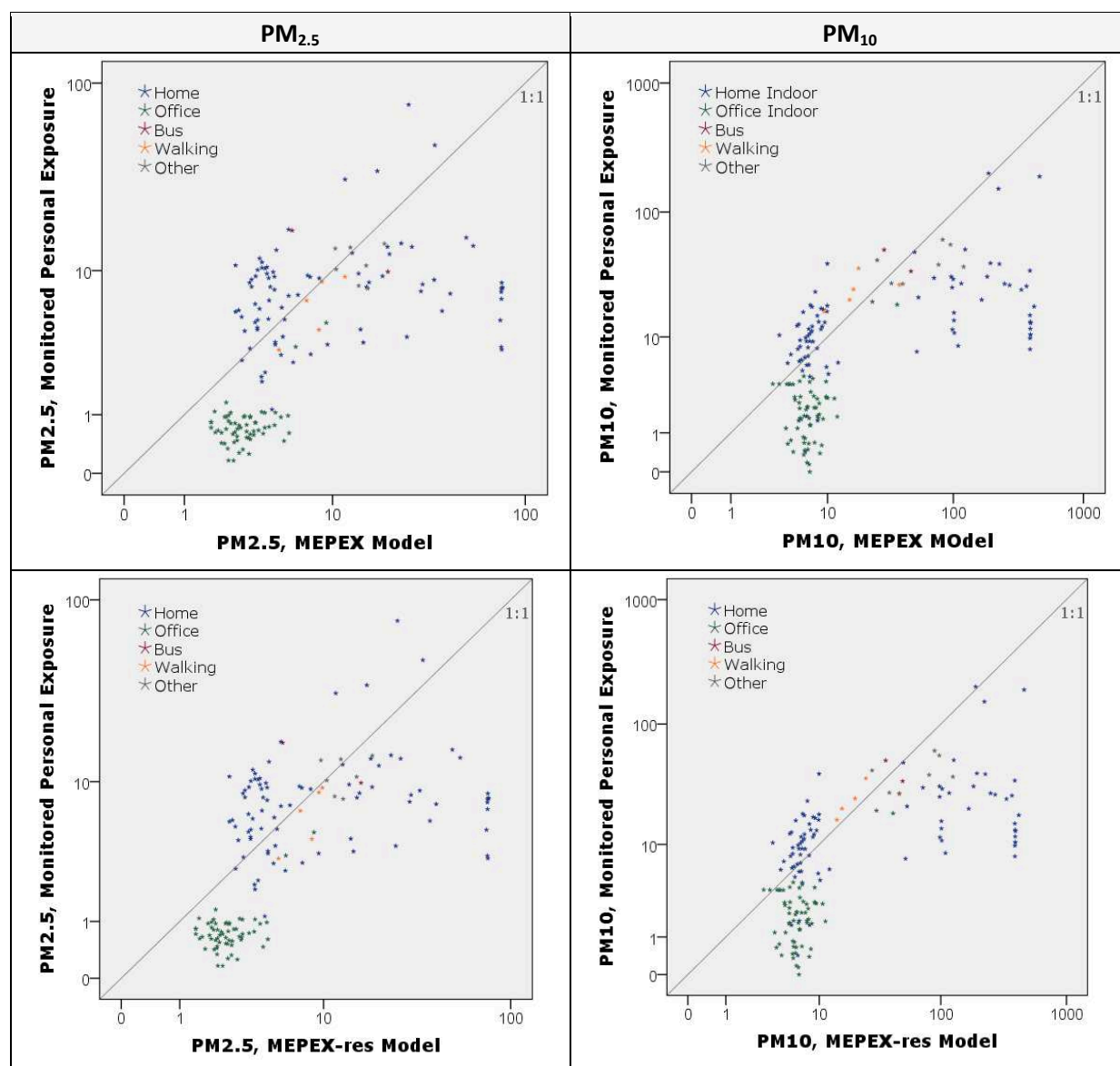


Figure 56: Modelled against monitored PM_{2.5} and PM₁₀ concentrations (both in $\mu\text{g}/\text{m}^3$) for hourly concentrations of personal exposure collected by individuals B, C, and D, specified by main visited microenvironment, for the MEPEX model (top) and the MEPEX-res model (bottom). Both X and Y axes are presented in log scale.

6.6. Model Application

In order to show the potential to predict personal exposure at a population level, the MEPEX-res model was applied for predictions of a study population in London. The personal exposure for a population of 6476 individuals was calculated for 2010 using the MEPEX-res model and compared to

ambient predictions at people's home locations. The application is intended to show the potential to predict personal exposure for a whole population while including ME differences. Simplified virtual time-activities are calculated for this purpose. Time-activities chosen for this demonstration are kept very simple, but could be refined according to study aim and purpose for future applications.

Data Preparation

Addresses for 6476 adult individuals were randomly selected within the boundaries of Greater London. PM_{2.5} concentrations of address locations for year 2010 were predicted with the ESCAPE-LUR model and provided for this analysis by colleagues from the ESCAPE study (for data protection, no exact locations were disclosed). These predictions were used as an input for the MEPEX-res model, as well as for the comparison of modelled estimates.

Real time-activity for individuals of the study population was not known. Simplified virtual time-activities were therefore constructed from time-activity survey data of the 'national time-use survey 2000' and the 'London travel demand survey 2011'. A limitation in this context was that not all data underlying the constructed time-activities was available for year 2010. The 'national time-use survey' 2000 (NTUS) is one of the most detailed time-activity surveys conducted for the UK, which collected time-activities for 20,981 individuals in 10-minute steps including 3164 different variables. The diary data was provided online by the UK data archive (<http://www.data-archive.ac.uk>, accessed: 19th Nov. 2009). Daily average times of sleeping, cooking, and in-transport have been calculated from the NTUS for this application.

The London travel demand survey (LTDS) is conducted annually to provide detailed information on travel behaviour of people in London. The LTUS 2011 provided data on the share of different transport modes for this application for year 2010 (Transport for London 2011). The survey included about 8,000 interviews.

Construction of Virtual Time-activities

In order to construct simple time-activity profiles for the study population, several assumptions were made:

- Individuals are either full-time employed or not working,
- All days of the year (2010) are working days,
- Total daily travel time of individuals is independent of mode of transport,
- The study population was not influenced by ETS (Environmental Tobacco Smoke).

The NTUS data was prepared for the extraction of information by selecting adult diaries from people living in the region of London with less than 90 minutes of missing entries (indicated as most suitable for analyses by the NTUS). The remaining dataset contained 16,345 diaries. In order to construct time activities for the study population it was distinguished between two main groups of people: working and not working. The employment rate of the adult population of London is 61.7 %, according to figures from the National Office of Statistics (<http://www.statistics.gov.uk>, dataset: regionalemploymentbyage_tcm77-359107.xls, accessed 1st May 2014) for 2013. In accordance to this statistic 61.7% of the study population were randomly sampled to represent working individuals. Only less than 1000 diaries of the NTUS for London were conducted by people who were in full-time employment. Average hours worked were therefore extracted from the National Office of Statistics (<http://www.statistics.gov.uk>, dataset: labou_tcm77-254358.xls, accessed 1st May) for April 2010, which was given as an average of 7.5 hours per working day. This average was rounded down to 7 working hours per day (not rounded up out of consideration for people with less than full-time employment contracts, who were otherwise neglected in this application). Time-activities for the study population were then extracted from the NTUS for:

- Average sleeping time: 514.4min or 8.53 hours, rounded to 9 hours,
- Average cooking time: 45 minutes,
- Average time in transport: 69 minutes (calculated both for employed and unemployed separately), rounded to 1 hour.

Selection of transport mode for the time spent in transport was randomly sampled for the study population according to the distribution of transport mode shares given by the LTDS. The LTDS data on mode shares per trip states that 12.8% of trips in London were undertaken using railway transport (including underground and overground), 15.2% were undertaken by bus, 30% by foot, 39.3% by car (passenger or driver), and 2.5% of trips were undertaken using motorbike or bicycle. Additional information of house volume was required for the application of an MB model for times at home. The average usable floor size of homes in England is given by the English Housing Stock Report 2008 as 91m² (Department for Communities and Local Government 2010), which was used as a guide size for this application. The ceiling height was estimated at 2.50m, providing an average home volume of 227.5m³.

MEs	Hours	
	Employed	Not Working
At home, sleeping	9h	9h
At home, active	5h	12h
At home, cooking	1h, incl. 45min of cooking	1h, incl. 45min of cooking
At home, hour after cooking	1h, calculated from cooking hour	1h, calculated from cooking hour
Work	7h	-
In-transport (randomly sampled mode)	1h	1h

Table 61: Hours spent in each microenvironment for virtual time-activity profiles of employed and not working individuals.

Final constructed time-activity profiles for the study population are presented in Table 61. The different MEs are modelled according to the previously described MEPEX-res model with times at home modelled by the MB model, times in transport adjusted by different mode to background ratios and time at work adjusted by an in-office to ambient ratio.

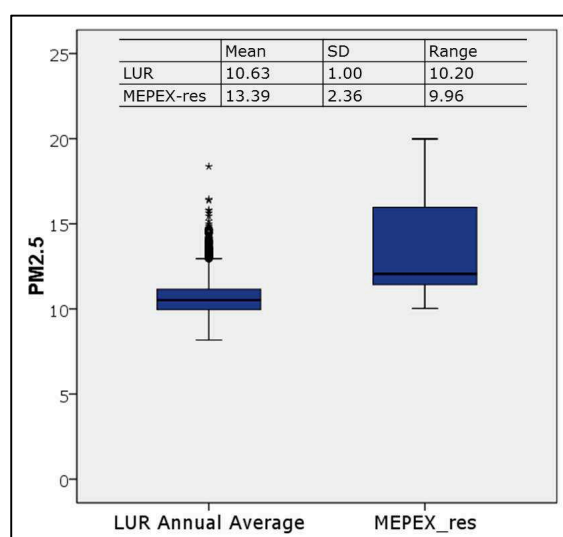


Figure 57: Personal exposure for a study population in London calculated using the MEPEX-res model in comparison to predictions of ambient concentrations at the home addresses for PM_{2.5} (both in µg/m³). Basic statistics are added in the graph.

Results

Results for personal exposure calculated by the MEPEX-res model, as well as by predictions of ambient concentrations at the home addresses are presented in Figure 57. For PM_{2.5} predictions of the MEPEX-res model show a mean concentration of 13.39µg/m³, an SD of 2.36µg/m³ and a total

range of $9.96\mu\text{g}/\text{m}^3$. Predictions based only on ambient estimates are in comparison lower and show less variability with a mean of $1.63\mu\text{g}/\text{m}^3$, an SD of $1\mu\text{g}/\text{m}^3$ and a range of $10.20\mu\text{g}/\text{m}^3$. A discussion of the results is included in the following section.

6.7. Discussion

In this chapter the MEPEX model was build, evaluated and compared to other methods of personal exposure prediction. Performance results for the compared modelling approaches show clear differences when applied to predict exposure for individual A.

6.7.1. MEPEX Model and Model Components: Evaluation of Hourly Predictions

Evidence of health effects after very short exposure to particles is increasingly recognised and predictions at an hourly level are considered an important part of exposure assessment. In addition, better hourly predictions could improve predictions for averaged exposure of longer time periods. Compared to other approaches, the MEPEX model could provide an improved tool for applications investigating sub-daily exposure effects. In the following section, the performance of the MEPEX model and its components for hourly predictions of individual A are discussed.

For hourly $\text{PM}_{2.5}$ predictions the model showed generally weak results for model performance parameters, which could be improved by excluding two outliers. IA results of model performance parameters for hourly PM_{10} concentrations showed a good model fit ($\text{IA} > 0.5$). The error (NMSE) was however comparably high and the model showed general overprediction.

The high error of the hourly estimates is to a large degree related to the high variability within each ME and the difficulty of the model to reflect the variability with few ME parameters. Results presented in chapter 3, as well as findings from the literature have shown that exposure variability within a day is mainly driven by differences between MEs and ME situations. These differences are represented in the MEPEX model by one or, for the home indoor ME, several parameters. Some of these MEs or ME situations show very high variability, as for example exposure during cooking or exposure in buses. The variability within each ME can only be reflected to a small extent by the parameters used in the MEPEX model. MEs and ME situations with high variability cause as such a high degree of uncertainty for predictions by the MEPEX model, in turn resulting in high error (NMSE).

Compared to methods, which include less parameters, the MEPEX model showed better model performance results for the prediction of hourly concentrations. For $PM_{2.5}$, R^2 and IA show the highest results in comparison to all other methods with 0.113 and 0.404 respectively. For PM_{10} , R^2 and IA results are as well higher compared to other methods with 0.210 and 0.629 respectively.

In order to understand the strengths and weaknesses of the MEPEX model it is important to evaluate the performance of the different ME components. Each of the main visited MEs was therefore evaluated separately. Several observations and conclusions are made:

- For the home indoor ME, results for model performance parameters were weak for $PM_{2.5}$, but improved after removal of the two cooking related outliers. For PM_{10} , predictions showed better performance with a good model fit ($IA > 0.5$). In comparison to using monitored outdoor concentrations (chapter 5), the MEPEX model (based on LUR-adj predictions for home outdoors) showed similar, and only slightly lower results for model performance parameters. As presented in chapter 5, the MB model showed difficulties reflecting the full variability of the monitored indoor concentrations, resulting in high error (NMSE). Over- and underpredictions of model parameters for different source related situations may in addition indicate poor parameterisation (definition of parameters).
- The office ME was substantially underpredicted by more than a factor of 2 for both $PM_{2.5}$ and PM_{10} . The adjustment applied for $PM_{2.5}$ was based on a study by Hänninen et al. (2005) including 94 buildings in Finland. The monitored concentrations from that study may have included naturally ventilated office, which could explain some of the observed overprediction. For PM_{10} , the applied ratio is only based on concentrations of two offices in Singapore, which may not have been representative for the ME (Gupta & Cheong 2007). Close to good model fit and a comparably low error observed for predictions indicate that contrary to home indoor exposure, variability of exposure inside offices could be predicted well by a simple ratio.
- For the cycling commute, predictions with the MEPEX model show medium to high results for R^2 and IA, with good model fit for both $PM_{2.5}$ and PM_{10} . However, estimates underpredicted cycling exposure considerably. The cycling ratio evidently did not reflect the full difference between estimated roadside concentrations and the cyclist's exposure. A likely explanation for the underprediction of the $PM_{2.5}$ concentrations is the lack of adjustment for vertical displacement between sidewalk concentrations and concentrations at monitoring stations, which underlie the ambient model. For PM_{10} , the sample size for the calculation of the cycling to sidewalk adjustment was small, based only on eight repeated journeys. The concentrations underlying the ratio may therefore have not been

representative for the difference between cycling and walking exposure along the route. For future applications, further validation of the ratios and possible development and combination with an adjustment for vertical displacement could be considered.

- Contrary to the cycling adjustment, the in-bus adjustment overpredicted exposure in the bus for individual A. The in-bus to sidewalk ratio was calculated by combining in-bus to cycling and cycling to walking ratios. The cycling to walking ratios have shown to underpredict exposure. This suggests an extreme overprediction of the in-bus to cycling ratios. In chapter 4 the variability along the routes and in between journeys has been evaluated for the dataset which was used for the calculation of the in-bus adjustments. No clear outliers could be detected which would explain the detected overprediction in this application. Another study by McNabola et al. (2008) calculated even higher in-bus to cycling ratios for PM_{2.5} (1.38 and 3.21 for different routes) compared to ratios calculated for this thesis, which would have resulted in an even more pronounced overprediction. Generally in-bus concentrations are highly variable (see chapter 2) with variability depending on a multitude of factors, such as number of passengers, window openings. Predictions of in-bus exposure using a simple ratio cannot reflect this variability, which may have influenced the accuracy of the predictions.

6.7.2. MEPEX Model: Evaluation of Daily Predictions

Daily exposure estimates have a much broader range of applications in the epidemiological context compared to hourly exposure estimates, as many health effects are recorded at a daily level for large population datasets (e.g. mortality or cardiac arrests).

Compared to hourly estimates, results for model performance parameters for the MEPEX model were improved for daily exposure predictions both for PM_{2.5} and PM₁₀. For time-weighted daily predictions of PM_{2.5} the model performance parameters showed good results with a moderate R² (R² = 0.439) and a good model fit (IA > 0.5). However, estimates generally overpredicted. For PM₁₀ the daily predictions also showed generally good model performance with a moderate R² (R² = 0.403) and a good model fit with IA well above 0.5 and a comparably low error (NMSE < 1). Overprediction was again observed for the modelled concentrations. The variability between and within different MEs and ME situations is only reflected to a small degree in concentration differences between days. The exact prediction of each ME or ME situation has as such less influence on the exposure estimates. The error (in form of the NMSE) is therefore much lower compared to hourly estimates and the R² shows higher linear agreement between modelled and monitored concentrations. The

comparison to other approaches for exposure modelling showed that at a daily level the detailed reflection of MEs in the MEPEX model results in improved exposure predictions compared to less detailed approaches. The MEPEX model can as such provide a tool for improved daily exposure predictions in future applications.

6.7.3. Evaluation of Predictions by Alternative Methods of Exposure Assessment

NEAR and AVERAGE method

Apart from the MEPEX model, other methods have been applied to predict personal exposure of individual A. The results clearly show that all methods based only on ambient concentrations perform generally less well compared to methods including ME models or adjustments both for hourly, as well as for daily concentrations of PM_{2.5} and PM₁₀.

Using the nearest monitoring station to a home address (NEAR) or an average of concentrations at monitoring stations (AVERAGE) in the area to predict personal exposure is one of the most commonly applied approximations for exposure in epidemiological studies (see chapter 2). The methods have however been criticised, as often only a relatively small part of actual personal exposure is reflected by these estimates, which leads to exposure misclassification for a large part of the study population (Avery et al. 2010; Özkaynak et al. 2013; Wilson & Brauer 2006). Results from this thesis also show that estimates based on the NEAR or the AVERAGE method perform weakly for exposure predictions of individual A.

Daily predictions result in better model performance parameters compared to results for hourly estimates. Both for daily PM_{2.5} and for daily PM₁₀, predictions the AVERAGE method showed slightly higher IA results compared to the ME-h model. Results for R² for the AVEARGE method were however lower than results for the ME-h model for both particle size fractions. In general, the results for daily estimates using the AVERAGE or NEAR method remained below results from models which included ME differences. This shows the potential of personal exposure models for improved exposure predictions and the reduction of exposure misclassification, compared to estimates based only on ambient monitoring concentrations.

AMB-h and AMB-ta model

Predictions of the AMB-h model and the AMB-ta model are both based directly on predictions of the LUR-adj model without ME adjustments. The AMB-h model predicts exposure using modelled ambient concentration for the home location. An approach which is commonly applied in large-scale epidemiological studies (Pedersen et al. 2013; Wang 2013; Gehring et al. 2013; Beelen, Raaschou-Nielsen, et al. 2014). The AMB-ta model uses the LUR-adj model to predict different locations visited during the day. Spatial differences of particle concentrations are widely recognised (Eeftens, Tsai, et al. 2012; Briggs 2007). The comparison between results of the AMB-ta model and the AMB-h model can show to what extent including individual daily movements would make a difference to personal exposure predictions, compared to predictions based on home location only. For hourly predictions, results for model performance parameters are better for predictions by the AMB-ta model compared to estimates by the AMB-h model, both for $PM_{2.5}$ and for PM_{10} . This may indicate that the reflection of different ambient concentrations of locations visited during the day could improve personal exposure models. For daily predictions of $PM_{2.5}$ however, the model performance results for the AMB-ta model are lower compared to results from the AMB-h model. For daily predictions of PM_{10} , results of R^2 and IA differ between the two models only by a small margin. The small difference between models for the results of daily predictions may reflect the fact that the time-activity on different working days is similar, and broadly the same locations were visited on different days. In comparison to other methods, results are similar compared to the NEAR and the AVERAGE method. Compared to models, which include ME differences, results for R^2 and IA are invariably lower. This indicates that the addition of ME adjustments to the predictions of ambient concentrations resulted in a clear improvement of model performance, which in turn has the potential to reduce exposure misclassification.

ME-h and ME-o model

With model ME-h the impact of using a simple I/O ratio instead of an MB model for home indoor MEs was tested. Results of the model performance parameters clearly show a substantial reduction of model performance for the ME-h model compared to the MEPEX model. This indicates that the choice of the MB approach as an indoor model is of high importance for the overall model performance.

For model ME-h the office ME was predicted using a home I/O ratio instead of an I/O ratio calculated for offices. Using a home I/O ratio results in an increase of the exposure overprediction for times at the office and clearly reduced results for performance parameters, both for $PM_{2.5}$ and for PM_{10} . The

use of a specific ratio for in-office exposure as such has an important influence on the performance of the personal exposure model.

MEPEX-res model

The MEPEX-res model used concentrations at the home location as a basis for exposure for all MEs. Predictions of exposure for individual A showed similar results for model performance parameters of the MEPEX-res model compared to the MEPEX model, both for $PM_{2.5}$ and for PM_{10} . The lack of spatial differences of the ambient concentration has in comparison to differences between MEs little effect on the model. It should be noted that on most days only relatively few different MEs away from home were visited by individual A. Time-activity profiles including more locations may have resulted in larger variability of ambient concentrations for predictions of the MEPEX model. This could have increased differences between the MEPEX and the MEPEX-res model. In addition, all MEs away from home (apart from 'other indoor') were adjusted using ratios (office, bus, and bicycle). Temporal variability of the MEPEX-res model for these locations is dependent on temporal variability of the LUR-adj model, which is the same for all locations in the study area. A different type of model for ambient concentration predictions, such as a dispersion model, may therefore have resulted in larger differences between the MEPEX and the MEPEX-res model.

Compared to the MEPEX model, the MEPEX-res model may provide a simpler and more applicable alternative. The MEPEX-res model requires less processing time and input data, as only the prediction of ambient concentration at one location is required per individual.

6.7.4. Transferability and Applicability of the MEPEX and MEPEX-res Model

Transferability of the MEPEX and MEPEX-res model to predict exposure for different individuals

The MEPEX and MEPEX-res models were applied to predict the exposure of individuals B, C, and D to evaluate transferability of the models to other individuals, apart from individual A. Results for model performance parameters for the two models were very similar, confirming that for future applications the MEPEX-res model may provide an alternative to the MEPEX model.

Application of the models to predict hourly exposure for individual B, C, and D showed mixed results. Predictions for individual B showed very weak performance results. Predictions for individual D on

the other hand resulted in generally better performance compared to predictions of individual A. Results for individual B showed high overprediction for some hours spent at home indoors, which may indicate a problem of the MB model to predict concentrations in small homes. Results for individual D would indicate that the MEPEX or MEPEX-res model can predict the exposure of some other individuals well. Further validation of the models is however required to answer how well the MEPEX or the MEPEX-res model would predict for different individuals of a population.

Evaluation of the potential for model applications

Calculation of personal exposure for PM_{2.5} with the MEPEX-res model was undertaken for a population in London as a demonstration for model applicability. Results show a higher mean by 2.76µg/m³ compared to predicted home ambient concentrations. This difference between mean concentrations would translate into a noticeable impact on health effects. Schwartz et al. (1996) associated a 10µg/m³ increase in PM_{2.5} with a 1.5% increase in total daily mortality. An increase of PM_{2.5} concentrations of 2.76µg/m³ would as such be associated with an increase of mortality of 0.41%. This result highlights the potential impact a ME refined model, such as the MEPEX-res model, could have for studies that investigate the health impact of exposure in a population. It should however be noted that the application was based on very simple virtual time-activity profiles, which can only to a small degree reflect the actual time-activity differences in the population.

The increased variability of the results for the MEPEX-res model in comparison to the predictions based on home ambient concentrations is another consideration. A very simple and static approach was taken for the time-activity profiles of the application. A more dynamic approach (e.g. as undertaken by the EXPOLIS study (Kruize et al. 2003; Hänninen 2005a)) would most likely have resulted in even higher variability, as a large range of different time-activity profiles would have been sampled. Increased exposure variability in a population has important implications for the health assessment of a population (Dons, Van Poppel, Kochan, et al. 2014). A large range of exposure would mean that some individuals can be expected to have a much higher risk of potential health effects than others.

In this thesis population groups were divided into full-time workers and home-makers. More differentiated population groups could be distinguished in future applications, according to study aims and objectives. Population groups have been differentiated for example for the EXPOLIS study into children, adults – working or studying, adults – not working or studying, and elderly (Kruize et al. 2003; Hänninen 2005a). In a study by Dimitroulopoulou et al. (2001a) home-makers, schoolchildren and office workers were distinguished. In addition, different possible time-activity profiles could be

calculated for each population group to reflect a more realistic picture of the differences between individuals, as for example undertaken by Dons, Van Poppel, Kochan, et al. (2014). Sampling techniques (e.g. random sampling, or Latin Hypercube technique, see chapter 2), could then be applied to sample time-activities for the population.

For future applications of the MEPEX model, the locations of individuals visited throughout the day are a further requirement. For smaller epidemiological studies, routes can be recorded for a study population using GPS. In addition, GIS-based methods offer the possibility to model routes, such as commutes, at a large scale. Routes have for example been calculated by the FEATHERS model for the population of Flanders, Belgium, which were then used in the development of a personal exposure model by Dons, Van Poppel, Kochan, et al. (2014).

In summary, the application of a ME refined model, such as the MEPEX-res model or the MEPEX model can be undertaken using freely available data on time-activities, as well as existing methods of time-activity and route modelling for the calculation of exposure at a population level.

6.7.5. General Limitations for the Evaluation of Model Performance

Several general limitations apply to the comparison of modelled and monitored exposure, as undertaken in this chapter.

First, the models were mostly applied and compared for only one person. The concentrations measured for this individual's personal exposure may however not be representative for others. Only limited data was available to test transferability for predictions of other individuals. Differences between individuals could for example lie in different habits (e.g. cooking behaviour), different time-activity profiles (home-maker instead of full-time worker), or MEs visited.

Another limitation lies within the measurements themselves. All measurements are subject to measurement error. The Osiris monitors used for the collection of personal exposure concentrations were calibrated to the EU reference method to reduce initial measurement error. The calibration can however only reduce uncertainty about the measurements, some potential for measurement error however remains.

6.7.6. Reflection about the MEPEX and MEPEX-res Model Performance in Reference to Existing Literature

Little validation of personal exposure models has been undertaken up to date by existing studies. In fact Dons, Van Poppel, Kochan, et al. (2014), mentioned that no validation of personal exposure models had up to this point been undertaken. As such, the application of the MEPEX model and modified model versions provides one of the first evaluations of the performance of personal exposure models.

The study by Dons, Van Poppel, Kochan, et al. (2014) provides validation of a developed personal exposure model for black carbon including 54 individuals which were each monitored for one week. The validation can only provide little information as a comparison to results for this thesis however, as the validation is undertaken for black carbon, not for $PM_{2.5}$ or PM_{10} . Dons, Van Poppel, Kochan, et al. (2014) concluded from their findings however that the developed AB^2C model estimated average personal exposure slightly more accurately compared to ambient concentrations, which were predicted for home areas. This confirms findings from this thesis, namely: that personal exposure models have the ability to reduce exposure misclassification compared to exposure predictions based only on ambient concentrations.

Only one other study by Gariazzo et al. (2014) undertook validation of a personal exposure model for $PM_{2.5}$. The monitored concentrations were by the author however considered to be subject to substantial measurement error. Conclusions or a potential comparison to results from this thesis can therefore not be derived from the results.

The results from this thesis as such provide some of the first quantifiable evidence that a personal exposure model could improve predictions of particle exposure estimates compared to commonly applied methods, based only on ambient particle concentrations.

7. Discussion

7.1. Thesis Summary

The motivation for this thesis was to improve the knowledge about models for exposure of individuals to particles. In reference to this motivation, the aims of this thesis are as follows:

- to build an integrated model for personal exposure assessment taking into account the temporal and spatial variability of ambient particulate matter, as well as microenvironments.
- to evaluate the performance of different approaches for personal exposure assessment, comparing varying levels of model sophistication.

In this thesis a personal exposure model was developed. In a first step literature was reviewed to better understand existing approaches and knowledge in the field. Then, data of ambient, personal and microenvironmental $PM_{2.5}$ and PM_{10} concentrations was collected in several air pollution monitoring campaigns. The collected data was later used in the development and evaluation of a personal exposure model, as well as its model components. In a next step the components of a personal exposure model were developed or selected. The development of a land-use regression model for the prediction of ambient concentrations was described. Methods for temporal adjustments of the land-use regression model were evaluated and the adjusted land-use regression model was validated. A set of microenvironmental adjustments, models, and estimates was compiled to alter ambient concentrations according to microenvironmental context. For the adjustment of transport microenvironments, some adjustments were calculated from collected monitoring concentrations, other adjustments and estimates were selected from the literature. Models for indoor concentrations were in addition applied and compared for predictions of concentrations at a home in London, in order to select suitable approaches for the integration into the personal exposure model.

The personal exposure model components: model for ambient concentrations, and microenvironmental models were integrated into the MEPEX (MicroEnvironmental Personal EXposure) model. The MEPEX model was applied to predict monitored personal exposure for individuals in London. Model variations were built by leaving out or altering model components, following approaches used in the literature. The performance of different methods was then compared for predictions of personal exposure for an individual in London. Transferability to other individuals and applicability of the model was furthermore evaluated.

Results of the personal exposure models, as well as model components have been discussed as part of each respective chapter. Below, the key findings, as well as main limitations are summarised,

followed by an outlook into possible future developments and applications. Lastly, final conclusions are presented.

7.2. Key Findings

Key findings of this thesis are summarised in the following section.

Model for Ambient Concentrations

A temporally adjusted land-use regression model was developed and validated for predictions of ambient concentrations with high spatial and temporal resolution. Model development of a land-use regression model was undertaken as part of the ESCAPE study. Different approaches of temporal adjustments for land-use regression models exist in the literature. The approaches have so far never been compared. A comparison of different methods undertaken as part of this thesis can as such be used as a guideline and reference for future studies. The comparison of different approaches to temporal adjustments showed that:

- A. for the majority of tested locations the average concentration from several background monitoring stations showed higher correlations with concentrations at other locations, compared to concentrations from the nearest background monitoring station.
- B. Temporal adjustments using concentrations aggregated to reflect temporal patterns (such as time-of-day, day-of-week and season) only explained a small amount of temporal variability for hourly, daily, or weekly concentrations at locations in the study area. Potential for transferability of the adjustment to another year or to other locations was as well limited by generally weak correlations with monitored concentrations. Using a temporal adjustment that directly applied concentration differences recorded at air pollution monitoring stations in contrast resulted for the same locations invariably in higher Pearson's correlations.

Validation of the temporally adjusted land-use regression model showed good model fit ($IA > 0.5$) and low error ($NMSE < 1$) for predictions of $PM_{2.5}$ and PM_{10} both at roadside and background locations in London, for hourly, daily, and weekly estimates. To the authors knowledge no direct validation of short-term estimates by a temporally adjusted land-use regression model has been undertaken until today. The results from this thesis as such provide important information on the performance of these models for future applications.

Estimates calculated with the temporally adjusted land-use regression model produced better results for model performance parameters in comparison to a well-established dispersion model

(ADMS-urban) for PM₁₀ predictions at the majority of locations in London (n = 26) for hourly, daily, and weekly estimates. It should however be noted that the temporal adjustment uses monitoring data to apply temporal variability. The evaluation of the land-use regression predictions therefore compares the variability between two monitored concentrations, which may make this an uneven comparison to the fully modelled ADMS-urban predictions.

The lack of a comparison between temporally adjusted land-use regression models and other models, such as dispersion models, has been mentioned before by Hoek et al. (2008). The comparison undertaken by this thesis between a temporally adjusted land-use regression model and a dispersion model provides such a comparison. The results indicate that the temporally adjusted land-use regression model is a good alternative to dispersion models.

Transport Microenvironments

Adjustments of transport microenvironments for PM_{2.5} and PM₁₀ were developed as follows: roadside to cycling and roadside to in-bus adjustments, as well as adjustments to background concentrations for walking, cycling, and in-bus exposure.

Predicting cycling exposure with the roadside to cycling adjustments showed good model fit (IA > 0.5) and medium to high linear agreement with concentrations monitored during cycling commutes both for PM_{2.5} ($R^2 = 0.852$) and for PM₁₀ ($R^2 = 0.486$). The adjustment for the horizontal displacement for cyclists' exposure in traffic has so far not been applied in other personal exposure models. Results for the prediction of cycling commutes indicate that the adjustment could correct for some of the concentration displacement.

Predictions of in-bus exposure during commutes, modelled with the in-bus adjustment showed a good model fit for PM_{2.5} (IA > 0.5), but not for PM₁₀. For both size fractions, R^2 remained low (PM_{2.5}: $R^2 = 0.359$, PM₁₀: $R^2 = 0.050$), and the model generally overpredicted exposure. Only one other study, by Gerharz et al. (2009) has implemented a specific adjustment for in-bus exposure into a personal exposure model. The applied ratio for that model was however calculated between in-bus exposure and concentrations at a nearby roadside station. Ratios calculated as part of this thesis are based on simultaneous measurements, which much better reflects the direct difference between concentrations inside and outside the bus. Several studies have used adjustments for in-car exposure to predict exposure inside buses for the development of a personal exposure model (Borrego et al. 2006; Briggs et al. 2005; Gulliver & Briggs 2005). In-bus exposure is however different from exposure inside cars. Buses for example regularly open their doors, which alters the degree of infiltration and buses show much higher passenger movement resulting in higher resuspension. The

developed in-bus adjustments can as such better reflect exposure inside buses compared to other approaches applied until today.

Models for indoor concentrations

The performance comparison of different models for predictions of hourly indoor $PM_{2.5}$ and PM_{10} concentrations (indoor-outdoor ratio, regression models, and mass-balance models) for a home in London showed:

- A. Predictions of all models resulted in generally weak results for model performance parameters for hourly predictions of a home in London both for $PM_{2.5}$ (all models: $R^2 < 0.52$, $IA < 0.26$, $NMSE > 8.43$) and for PM_{10} (all models: $R^2 < 0.18$, $IA < 0.59$, $NMSE > 7.12$)
- B. The performance of models for residential indoor environments based only on ambient concentrations showed lower R^2 and IA , as well as higher $NMSE$, compared to approaches, which distinguished different situations indoors (e.g. cooking or movement of occupants), both for $PM_{2.5}$, as well as for PM_{10} .
- C. Regression models for residential indoor microenvironments, as applied in this thesis, may provide an alternative to other methods. Weak performance for predictions of monitored concentrations in this thesis however indicate that further validation or a redevelopment with a bigger dataset may be beneficial.
- D. Best performance for predictions of home indoor concentrations were recorded for a mass-balance model, which included cooking, 'active occupancy', and 'hour after cooking' as source parameters (model MB3b). For $PM_{2.5}$ results for performance parameters of model MB3b however showed only limited ability to predict concentrations indoors ($R^2 = 0.051$, $IA = 0.251$, $NMSE = 9.107$, $FB = -0.044$). For PM_{10} , the model showed good model fit ($IA = 0.584$), R^2 however remained low ($R^2 = 0.174$) and model error and overprediction were high ($NMSE = 8.108$, $FB = 0.520$).
- E. Two new parameters were calculated for the mass-balance model (MB3b) to adjust for 'active occupancy' (people at home and not sleeping) and for raised concentrations during the 'hour after cooking'. Including the newly developed parameters clearly increased results for R^2 and IA , and reduced error ($NMSE$) for both particle size fractions, compared to results calculated only with cooking as a source parameter.

The generally weak performance of all compared indoor models may indicate that none of the models can predict indoor concentration well. Several other possible reasons for the weak performance have been addressed in chapter 5 and include: skewed distribution of the monitored

dataset, possible measurement error, and the potential for unusual indoor concentrations in home A. Little is known about the performance of most of these models. In a study by Hänninen et al. (2004) it was however concluded that even after adjusting for all known influences to indoor concentrations, a large part of indoor PM remains unexplained.

Indoor-outdoor relationships have been investigated by a range of studies. Highly varying correlations between indoor and outdoor concentrations have been presented in chapter 2 with some correlations below 0.1 (both Pearson's correlation and Spearman's Rho). Results from this thesis for the performance of the indoor-outdoor ratio as such confirm that the use of a simple indoor to outdoor relationship can only reflect a small part of the concentration variability indoors. Weak results for model performance parameters of the indoor-outdoor ratio are therefore in accordance with findings from the literature.

Of the applied regression models, the REG-J2b model performed best, including outdoor concentrations, sleeping, sitting, walking, and cooking. Similar to findings from the application in this thesis, the original study showed low R^2 results for the validation of the model for fine PM. For PM_{10} original validation results showed a much higher R^2 ($R^2 > 0.5$), compared to results from this thesis, which may indicate that the model predicts for some homes better than for others.

Little is known from the literature about the performance of the MB models, as applied here (a one-compartment model with deterministic parameters). Most existing studies on MB models focus on the establishment of parameters or further developments of the mass-balance model (e.g. towards a multi-compartment approach, as by Dimitroulopoulou et al. (2006)). Mass-balance models, as applied in this thesis, are however frequently applied in the literature (Koutrakis et al. 1992; Gerharz et al. 2009; Hoek, Kos, et al. 2008; Hänninen et al. 2004; Ferro et al. 2004; Nazaroff 2004). The comparison and evaluation undertaken here can as such improve the understanding of these models for future applications. Despite the generally weak performance the comparison between different methods clearly showed best performance for the MB model when several parameters for indoor sources were included. The improvement of model performance with the added parameters for 'active occupancy' and 'hour after cooking' can provide a further direction for future applications.

MEPEX model

The MEPEX model, an integrated personal exposure model was built, including more detailed parameters for microenvironments and microenvironmental situations compared to most other models for personal exposure. Applied to predict personal exposure for an individual in London, the model showed good model fit ($IA > 0.5$) for hourly and daily predictions of PM_{10} , as well as for daily

PM_{2.5} predictions. Hourly PM_{2.5} predictions showed as well good model fit after the removal of two cooking related outliers. Compared to other, simpler approaches of personal exposure predictions, the MEPEX model showed some of the best results for model performance. R² results for daily predictions were moderate both for PM_{2.5} and PM₁₀ with 0.439 and 0.403 respectively. Results for IA showed good model fit both for PM_{2.5} (IA = 0.661) and for PM₁₀ (IA = 0.725). The model however overpredicted for both particle size fractions with an FB of 0.208 for PM_{2.5} and an FB of 0.451 for PM₁₀.

Few existing personal exposure models have included a model for ambient concentrations with both high temporal and spatial resolution. Only one other existing personal exposure model has in addition included detailed adjustments and models for different microenvironments. This other model was developed by Gerharz et al. (2009) as a pilot study to predict PM_{2.5} concentrations for 14 daily time-activity profiles in Muster, Germany. The study used a temporally adjusted dispersion model for ambient concentrations, as well as ratios to adjust for in-bus and in-car exposure. Indoor concentrations were modelled using a mass-balance model. Only the indoor model of this approach was compared to monitored concentrations. No comparison was undertaken by Gerharz et al. (2009) with monitored concentrations of personal exposure. As detailed in chapter 6, altogether very little validation has been undertaken for personal exposure models by other studies. The MEPEX model as such provides the first detailed approach to personal exposure modelling for which performance was evaluated using monitored personal exposure.

Models based only on ambient concentrations

Exposure estimates for individual A based only on ambient concentrations showed only weak linear agreement with hourly monitored personal concentrations. R² results for daily estimates of these approaches are in comparison higher, yet remain generally weak with R² ranging between 0.117 and 0.173 for PM_{2.5} and between 0.020 and 0.086 for PM₁₀. Results for IA are mostly also low, daily PM_{2.5} prediction using concentrations monitored at the nearest monitoring stations however showed an IA above 0.5, indicating good 'model' fit.

Results from this thesis clearly show that any approach using only ambient concentrations to predict personal exposure resulted in exposure misclassification for individual A. Nearest monitor, average of several monitoring stations in the area, as well as modelled concentrations at the home address are established methods for the prediction of exposure, especially in larger studies. The results from this thesis clearly show that for some individuals, these methods can only predict little of the

actually experienced short-term personal exposure. The integration of microenvironmental components into the models on the other hand clearly improved performance.

Importance of different indoor models for model performance

93% of our time is on average spent indoors with most of the time spent at home or at work (Schneider et al. 2004). Predictions of indoor microenvironments in a personal exposure model are therefore of particular importance. The comparison of different approaches could show that the use of different indoor models has an impact on the performance of the personal exposure model. To adjust personal exposure models for the home indoor microenvironment most existing studies have applied indoor-outdoor ratios (Dons, Van Poppel, Kochan, et al. 2014; Borrego et al. 2006; Borrego et al. 2009; Baklanov et al. 2007; Kousa, Kukkonen, et al. 2002; Gariazzo et al. 2014). The findings from this thesis however indicate that the use of a mass-balance model should be preferred, as the use of an indoor-outdoor ratio clearly reduces overall model performance for personal exposure predictions. Furthermore, results indicate that for exposure predictions of the office microenvironment, the use of an indoor-outdoor ratio based on concentrations measured in the office should be preferred over the use of a home indoor-outdoor ratio.

MEPEX-res model

The MEPEX-res model calculated personal exposure using microenvironmental adjustments based only on home ambient concentrations. Performance of the MEPEX-res model showed very similar results compared to the MEPEX model for estimates of exposure for individual A, as well as for exposure estimates of individuals B, C, and D. Including individual location differences had as such little impact on overall model performance.

In comparison to the MEPEX model the MEPEX-res model has fewer requirements on data and processing times, as no individual data on people's locations is required and ambient concentrations only need to be calculated for home addresses. The MEPEX-res model as such provides a simplified alternative to the MEPEX model, especially for applications including large populations.

7.2. Limitations

General Limitations

Several general limitations apply to the approach taken in this thesis:

The monitored concentrations underlying the model for ambient concentrations, as well as some of the microenvironmental parameters are based on fully EU reference equivalent concentrations (monitored by TEOM/FDMS and Harvard Impactor devices). Monitored concentrations for personal, indoor, outdoor, and transport microenvironments collected as part of this thesis were initially not reference equivalent, but have been calibrated to reference methods using equations developed by Walden et al. (2010) before analyses. The study by Walden et al. (2010) showed that calibrated PM₁₀ concentrations of Osiris monitors generally qualified for full reference equivalence (tested for a full dataset and several data subsets). Calibrated concentrations of PM_{2.5} however only qualified to be indicative of concentrations monitored by EU reference methods.

Qualification for EU reference equivalence is based on limits for the degree of accepted measurement uncertainty (using a combination of different parameters for measurement uncertainties for different data subsets, see also: EC Working Group on Guidance for the Demonstration of Equivalence 2010). Measurement uncertainty is the "... quantification of the doubt about the measurement." (Bell 1999). Methods calibrated to EU reference equivalence are commonly used and interpreted as a close approximation to the 'true' concentrations. For the UK FDMS/TEOM monitors, run by monitoring networks of the LAQN and the AURN, are for example calibrated to reference equivalence. UK health warnings for days of high PM concentrations, as well as calculation of exceedences of limit values are based on these calibrated concentrations, (<http://uk-air.defra.gov.uk/networks/network-info?view=aurn>, accessed: 28th May 2014; <http://www.londonair.org.uk/london/asp/publicdetails.asp?region=0>, accessed: 28th May 2014).

As a consequence, calibrated PM₁₀ concentrations from Osiris monitors are considered to be a close approximation to the 'true' concentrations. Calibrated PM_{2.5} concentrations however did not meet the full standard for reference equivalence and have in comparison higher measurement uncertainty. As a result a higher measurement error may underlie the respective monitored concentrations in this study. This could have resulted in increased differences between monitored and modelled concentrations. In turn this may have reduced general results of model performance for predictions of indoor concentrations and for predictions of personal exposure concentrations.

Data presented as part of this thesis only represents part of the initially collected datasets. In addition to the presented data, several pilot campaigns were undertaken to test for example options to carry monitoring devices. Furthermore, a considerable amount of data was lost due to

measurement failure of the Osiris devices (an estimated 35%). Specific reasons for this data loss are listed in Table 62.

Type of Data Loss	Consequence	Affected Data/ Campaign
1) Repeated malfunction of electronic connections between core monitor and heated inlet	Data recorded without heated inlet was removed from datasets	Transport monitoring campaign
2) Repeated malfunction of electronic connection between core monitor and power supply	No data recorded	Indoor-outdoor campaign
3) Malfunction of the internal clock of all Osiris devices causing the time stamp to lag by between 5 and 15 minutes per month	Data collected before recognition of the default was removed from datasets	Start of monitoring for individual/home A
4) Initial difficulties to set up monitor correctly	Monitor did not start or did not collect intended type of data	Start of monitoring for all four individuals and their homes
5) Housemate switched off the power at a socket used for monitor's power supply (once)	No data was collected	Indoor, home B
6) Battery ran out of power before the end of the monitoring day (3 times)	Data was not collected for the full time away from home	Personal exposure monitoring campaign
7) Sudden loss of data recorded by the GPS device	Monitoring data was removed from dataset, as start and end time of routes were unclear	Transport monitoring campaign

Table 62: Reasons for data loss during air pollution monitoring campaigns.

Most of the data loss or removal could not be attributed to a systematic error and occurred at random points in time. A slight bias towards the collection of personal exposure during the morning hours (e.g. morning commute was captured, but not evening commute) may have been caused by days during which the battery pack ran out of power while away from home. Furthermore, increased data loss was observed when individuals started to handle the devices. This in turn caused less problems for the more continuously monitored individual/home A, compared to the shorter monitoring periods planned for individuals/homes B, C, and D.

As a result of the data loss, some of the datasets collected for this thesis are smaller than initially intended, e.g. for the walking-cycling transport mode campaign. The concentrations of some of these smaller datasets may be less representative for the individual, situation, or ME, compared to the intended larger dataset.

Model for ambient concentrations

Several limitations apply to the adjusted land-use regression model, which have been detailed in chapter 4. One of the main limitations was that for the temporal adjustment of the land-use regression model, monitoring concentration data for the relevant area and time period of

application is required. For London, data from a large number of monitoring stations is however available, providing PM_{2.5} and PM₁₀ concentrations since the 1990s. Few temporal constraints for applications in London therefore exist. Monitoring networks for locations outside London are less dense (<http://uk-air.defra.gov.uk/interactive-map?network=aurn>).

Indoor microenvironments

The model selected for predictions of indoor concentrations (MB3b) in the MEPEX model showed generally only limited ability to predict PM_{2.5} concentrations indoors. For PM₁₀, the model showed good model fit (IA > 0.5), the model error however remained high. In addition, the model over-and underpredicted several model parameters, when applied to a home in London. It could be observed that for this application the parameter calculated for 'active occupancy' overpredicted for both size fraction. The cooking parameter, selected from the literature for PM₁₀, additionally overpredicted cooking related concentrations. The parameter for hour after cooking was dependent on the cooking parameters, and therefore increased overprediction for predictions for PM₁₀ concentrations. A general limitation of the comparison between different indoor models was that model performance in relation to ETS exposure could not be evaluated with the monitored indoor concentration.

The applied ratios for in-office adjustments are well below most ratios calculated for home indoor environments (see chapter 2), yet predictions for the in-office exposure of an individual in London (individual A) still showed overprediction by more than a factor of 2 both for PM_{2.5} and PM₁₀. The selected ratios therefore do not fully reflect the observed difference between ambient concentrations and in-office exposure.

Adjustments for Transport Microenvironments

The developed cycling to roadside ratio showed underprediction when applied to exposure during cycling commutes for individual A, which may be related to discrepancy between exposure on the sidewalk and at roadside monitoring stations due to a vertical particle displacement.

The in-bus exposure showed a good model fit for PM_{2.5}, but low R², as well as weak performance predictions of PM₁₀. Additionally the estimates overpredicted exposure. It is likely that in-bus concentrations are poorly reflected by a simple ratio. In-bus exposure is complex and studies have shown that, especially for coarse particles, influences from inside the bus significantly contribute to particle concentrations (Vijayan & Kumar 2010).

MEPEX Model

Over- and underprediction of different microenvironmental parameters and components can be observed for the model and better parameterisation of these components may have further improved predictions. It is however also a possibility that personal exposure collected for individual A is not representative of exposure in certain microenvironments or microenvironmental situations. Furthermore, time-activity patterns and specific microenvironments visited by individual A are only representative for some people. The importance given to certain microenvironments would be different for individuals who follow another time-activity pattern. Performance of the model to predict concentrations of personal exposure for three other people was tested, but datasets were too small to indicate how the model would work for different individuals of a population. The model was as such not fully validated for future applications. Before future applications of the MEPEX model may be considered therefore a larger validation study would be beneficial.

The performance of the MEPEX model was in this context only evaluated for hourly and daily predictions; some applications however require longer-term exposure estimates. It could be an interesting future development to examine the performance of the MEPEX model for longer time periods.

In this thesis the MEPEX model was developed for predictions of $PM_{2.5}$ and PM_{10} . Serious health effects have however as well been recorded for a range of other pollutants, such as for nitrogen oxides (NO_x), or sulphur dioxide (SO_2) (World Health Organisation 2006). Other pollutants generally show different physical properties (e.g. different distribution, different infiltration indoors) compared to $PM_{2.5}$ and PM_{10} . A direct application of the MEPEX model for other pollutants could therefore not be considered. The development of a personal exposure model, similar to the MEPEX model, may improve predictions of personal exposure for other pollutants.

A main limitation for applications of the MEPEX model is the requirement of information on individual's locations throughout the day. In particular route locations during commutes are usually not available for larger populations. GIS based methods have been used in the past to model commuter's routes for study populations (Briggs 2005). A further data requirement is time-activity patterns of the study population. As outlined in chapter 6, these can be modelled from existing time-use surveys.

Transferability of the MEPEX model in space and time is mainly limited by the model for ambient concentrations. The adjusted land-use regression model has been developed for London and surrounding areas. In terms of temporal transferability the model can be adjusted to predict all years for which background monitoring station concentrations are available (since the mid-1990s). The

microenvironmental adjustments and models are to a large extent considered transferable at least within the European context (similar home and transport configurations).

MEPEX-res Model

Model performance of the MEPEX-res model in comparison to the MEPEX model may be reduced if an individual spends many hours in locations with ambient concentrations different from concentrations at home outdoors. In addition, the similarity between the MEPEX and MEPEX-res model is most likely dependent on the fact that the LUR-adj model for ambient concentrations predicts the same temporal variability at all locations of the study area. Another model for ambient concentrations would most likely have shown more differences between model performance.

7.3. Future Applications and Outlook

The validated approach of the temporally adjusted land-use regression model can directly be applied to predict ambient concentrations with high-spatial and temporal accuracy. In addition, the result of the comparison between approaches for temporal adjustments could provide a useful guideline for the future development of temporal adjustments for land-use regression models. For simple estimates of personal exposure, home ambient concentrations can be predicted using the adjusted land-use regression model for study populations in and around London. Areas of application may for example include studies of pregnancy cohorts (trimesters) or studies investigating short-term health effects, such as asthma attacks or myocardial infarction. For applications in study areas away from London, either a validation of the model with local concentrations or a recalculation of the adjustments with data from local monitoring stations would be required. Furthermore, the application of a different land-use regression model can be considered for another area, temporally adjusted with local background monitoring data.

The developed adjustments for in-transport microenvironments can be directly integrated into personal exposure models. For the roadside to cycling adjustment a further development to calculate and combine a ratio adjusting for vertical displacement could be considered. An additional development could be made to adjust not only for cycling exposure, but also for the increased inhalation during cycling activities to calculate particle intake (Nazelle & Nieuwenhuijsen 2009).

The roadside to in-bus ratio can only be a crude estimate for the complex influences on exposure inside buses. A more complex modelling approach may be able to reflect the in-bus variability better and could be a direction for future research (Vijayan & Kumar 2010). Attempts to develop models for

in-bus exposure including for example passenger number have been made for several particle size fractions (Vijayan & Kumar 2010; Song et al. 2009), however not for $PM_{2.5}$ and PM_{10} . The development of a transferable model for in-bus exposure of $PM_{2.5}$ and PM_{10} for the inclusion in personal exposure models would however clearly benefit exposure estimates for in-bus exposures.

Existing literature has shown a clear need for more knowledge about exposure in railway transport microenvironments. This knowledge would be of crucial importance to improve parameterisation for personal exposure models, such as the MEPEX model.

Models for indoor home concentrations would clearly benefit from further research into using the regression modelling approach, which could provide an interesting alternative to mass-balance models. Furthermore, the parameters of the mass-balance model would benefit from further validation. Parameters for cooking source activities in particular have for this thesis shown a tendency to underpredict for $PM_{2.5}$ and overpredict for PM_{10} . In addition, the differentiation of the parameter for 'active occupancy' for example by number of people or type of movement may further improve indoor concentration estimates. It is important to notice that the applied static approach to parameters can never reflect the full concentration variability produced by the existing indoor sources. A possible future direction could therefore lie in an approach applied by the INDAIR study, for which a probability function was used to sample each parameter of the mass-balance model from a distribution (Dimitroulopoulou et al. 2006).

Little is known about exposure in modern office microenvironments. Further research and the establishment of I/O ratios which better reflect these environments could further improve the parameterisation of models, such as the MEPEX model.

The MEPEX model could directly be applied to predict individual exposure for a study population. The performance of the model was mostly evaluated for the prediction of personal exposure of one individual. A large validation study would therefore be the logical next step. A probabilistic approach to the selection of the parameters could be considered in which each parameter would be sampled from a distribution. Gerharz et al. (2009) suggests as well the application of a probabilistic approach as a potential future development for personal exposure modelling.

For future applications it should be considered that the calculation of the MEPEX model would require higher resources compared to simpler approaches, such as using concentrations from the nearest monitoring site. These requirements need to be weighed against the merits of the approach (see also: Briggs 2005). To a large degree these considerations depend on study design and aim. A small study for example investigating hourly exposure would clearly benefit from the application of the MEPEX model, as 1) for small study populations time-space-activity profiles can often be

collected using questionnaires and GPS devices, and 2) hourly exposure differences are particularly poorly reflected by ambient concentrations. The detailed approach of the MEPEX model provides in addition great potential for applications, which look into the differences between individual's exposures, such as implication of route choices during commutes on exposure and health effects.

For the application to larger populations, time-activity profiles cannot be collected individually. Information on time-activities for people in the UK has however been collected for example in form of the UK time-use survey. As presented in this thesis, this type of data can be used to build virtual time-activity patterns for a population by using a deterministic approach or by using probabilistic sampling. In addition to time-activity profiles, the MEPEX model also requires data on individual locations visited throughout the day. For the modelling of routes, GIS provide route modelling techniques which can for example model the shortest route between home and work locations along a road network. As an alternative, the MEPEX-res model could be applied, which requires only home address concentrations for each individual. The MEPEX or MEPEX-res model could as such be applied to predict exposure for large populations.

Following these considerations, potential applications (after further validation) for the MEPEX and MEPEX-res model mainly fall into three categories:

- 1) Small short-term predictions focusing on individual exposure (MEPEX model)
- 2) Comparative studies between population/lifestyle groups (MEPEX model)
- 3) Large-scale epidemiological studies (MEPEX or MEPEX-res model)

In the following paragraphs examples of potential MEPEX or MEPEX-res applications for these categories are discussed.

A study by Dons et al. (2011) provides an example for a small study which uses short-term estimates to assess health effects in school children. The study was conducted as part of the HEAPS (Health Effects of Air Pollution in Antwerp Schools) project and assessed the relationship between air pollution exposure (NO₂ and BC) with data collected in a biomonitoring study for 130 children. Concentrations measured at the school two hours before the medical examination were compared with measurements for oxidative stress, inflammation and cardiovascular markers. The MEPEX model could for similar studies provide exposure estimates of the study population for a range of hours leading up to the examination, including for example exposure estimates for the school commute. Using the MEPEX model in similar biomonitoring studies could therefore enhance the knowledge of the short-term effects of PM on the human body.

Short-term effects of PM exposure have as well been assessed for some specific health effects, such as myocardial infarction (MI). Peters et al. (2001) for example studied the onset of MI for 772

patients in the Boston area in relation to centrally measured PM_{2.5} concentrations two hours before the onset of an MI. Results show a significant relationship between increased PM_{2.5} concentrations and MI onset. Personal exposure differences experienced throughout the day are highly dependent on people's time activities and different MEs visited. The MEPEX model could be applied to predict exposures for similar studies, providing a much more refined estimate of personal exposure compared to centrally measured concentrations. Patient information for the study by Peters et al. (2001) had been collected during personal interviews. Information on time-activities and visited MEs of each patient could have additionally been collected during the interviews.

Several studies have conducted research that falls into the second category of potential applications: comparative studies between population/lifestyle groups. A study by Dons et al. (2011) has been mentioned above, for which exposure differences between partners were investigated. The couples were selected so that one of the partners was a home-maker, the other an office worker. The study by Dons et al. (2011) found significant differences in exposure between partners of eight couples using personal exposure monitoring data. A larger group of participants could be investigated using the MEPEX model for exposure predictions. The results of such a study could have significant implications for gender differences observed for some health endpoints (Schikowski et al. 2014).

Some studies have also been conducted on route choices of commuters (Hertel et al. 2008; Pattinson 2009). Exposure estimates for the routes have by these studies been based directly on estimates of ambient concentrations at roadsides. The MEPEX model could provide estimates adjusted for the horizontal displacement of in-traffic exposure and the analysis could furthermore be differentiated by modes of transport. Additionally, commuter exposure calculated by the MEPEX model could be put in context of overall daily personal exposure.

A large body of research has been undertaken which falls in the third category of potential applications: Large-scale epidemiological studies. These studies include both cohort and time-series studies that would be too large for the collection of individual scale information on time-activities.

For long-term exposure a large range of health effects was for example studied by the ESCAPE project, including lung function in children, cerebrovascular events, and natural-cause mortality (Gehring et al. 2013; Stafoggia et al. 2014; Beelen, Raaschou-Nielsen, et al. 2014). Individually calculated pregnancy trimester have been studied in connection to PM exposure by Pedersen et al. (2013) and by Slama et al. (2007). Large studies have as well been undertaken for short-term health effects, such as for ischemic stroke and mortality (Maynard et al. 2007; Wellenius et al. 2012; Pope III & Dockery 2006).

It has been shown as part of this thesis that the MEPEX model can improve hourly and daily exposure predictions for some individuals compared to methods traditionally used in time-series or cohort studies. The MEPEX and MEPEX-res models are as such particularly well suited to provide short-term exposure estimates at an individual level. Potential applications would for example include cohort studies on a large range of short-term health endpoints, including daily mortality, ischaemic stroke, MI, asthma attacks, as well as emergency room visits.

Long-term exposure is to some extent the average of repeated exposures during a daily routine. It has been shown in this thesis that the exposures experienced during a daily routine depend on specific MEs visited and may therefore substantially vary between individuals. The MEPEX and MEPEX-res model could provide a more realistic estimate for exposure variability within a population, compared to variability which is only based on home outdoor concentrations. This added variability would however depend on the approach taken for the modelling of population time-activities and the degree to which time-activity differences in the population are accounted for in the analysis. In addition, the prediction of long-term personal exposure has not been evaluated by this thesis and validation with long-term monitored personal exposure would be of particular importance. It should also be considered that for some health effects, such as several cancers whole-life exposure can be important. It would be very difficult to reconstruct time-activity profiles of a population for such extended time periods.

Additionally to the influence this thesis could have on the methods of future exposure assessment, results from this thesis could also have implications for future policies. The main focus of current air pollution policies is on ambient air pollution (Air Quality Expert Group 2012). It has been shown by several studies that personal exposure is only partially influenced by ambient concentrations (Avery et al. 2010; Özkaynak et al. 2013), which has been confirmed by results from this thesis.

This thesis has clearly shown the great importance of certain MEs and ME situations as an influence on personal exposure. Certain MEs and ME situations are typically revisited during each individual's daily routine. Personal exposure 'hot spots' can occur for times spent in MEs or ME situations which are associated with extremely high PM concentrations, such as during and after cooking or in certain transport MEs. For some people certain exposure 'hot spots' form part of their daily routine, which not only causes repeated short-term peak exposures, but also increasing their long-term average exposure.

Policies are currently aimed at reducing ambient background PM concentrations (Air Quality Expert Group 2012). While such a reduction would be beneficial, this focus ignores a great potential for the reduction of personal exposure by focusing more on certain MEs or ME situations. Possible areas of

future policies could for example lie in improved ventilation for kitchens or public transport microenvironments.

7.4. Final Conclusions

Results from this thesis show weak performance for traditional methods of exposure assessment, which base exposure on estimates of ambient concentrations for home locations, both for $PM_{2.5}$ and PM_{10} predictions. Findings as such confirmed existing concerns about these commonly applied methods to predict individual exposure well. The development of improved methods for exposure assessment towards more personalised exposure estimates is therefore necessary (see also Steinle et al. (2013)).

This thesis has contributed to the development of such a personal exposure model in several ways.

Firstly, a model for personal exposure assessment of $PM_{2.5}$ and PM_{10} , the MEPEX model, was built including ambient concentration estimates with high spatial and temporal accuracy, as well as detailed estimates and models for microenvironmental adjustments. Only one existing model by Gerharz et al. (2009) has up to now developed an approach to personal exposure modelling with a similar degree of detail. This model has however never been compared to monitored concentrations. Predictions of the MEPEX model for an individual in London show that for daily predictions the model can provide good estimates for personal exposure.

Secondly, for the integration into the personal exposure model several different components were developed, validated and compared. A temporally adjusted land-use regression model for $PM_{2.5}$ and PM_{10} has been built and validated, which can directly be applied to provide ambient predictions at high temporal and spatial resolution. Several ratios for in-transport microenvironments have been developed for the adjustment from ambient concentrations to in-transport exposure, which can be integrated as parameters into personal exposure models. Models for indoor microenvironments have been compared, revealing strengths and weaknesses of existing approaches, which provides crucial information for future research in that area. The comparison of the MEPEX model to less sophisticated personal exposure model variations has additionally revealed information on the importance of different model components. It has been shown that the selection of indoor models, both for residential and office locations is important. In addition, results revealed that in the context of this thesis the differences between visited geographic locations of an individual were of minor importance. With the MEPEX-res model this thesis provides an approach, which calculates microenvironmental differences based on ambient concentrations at the residential address. Similar

performance between MEPEX and MEPEX-res model could be observed, which indicates that the MEPEX-res model could provide a good alternative to the MEPEX model for future applications.

Thirdly, very few existing European studies have evaluated the performance of a personal exposure model for the prediction of monitored particle concentrations (Gariazzo et al. 2014; Dons, Van Poppel, Kochan, et al. 2014). Only one other study has applied a personal exposure model to predict $PM_{2.5}$ (Gariazzo et al. 2014). Results from that study however could not be compared due to measurement error. No study up to today has evaluated performance for PM_{10} . The results from this thesis are as such one of the first attempts to evaluate the performance of personal exposure models for particle concentrations. Compared to using estimates based only on ambient predictions, predictions by personal exposure models have clearly shown better model performance. This provides one of the first quantifiable results that the application of personal exposure models could improve exposure assessment. As a consequence using personal exposure models for exposure assessment could have the potential to reduce exposure misclassification.

For future applications the MEPEX model could provide improved estimates of individual exposure for short-term predictions of $PM_{2.5}$ and PM_{10} . The model is particularly well suited to predict exposure differences at a smaller scale, for which individual data can be acquired comparably easily. Another field for potential applications is the assessment of exposure differences between individuals, for which different behaviours or population groups are compared. These applications often require detailed short-term predictions of exposure estimates.

For larger scale applications considerations on data availability and processing times are more important. The MEPEX-res model may provide therefore a good alternative for large scale applications. The potential for an application of a similar personal exposure model (based on home ambient concentrations) has for example been demonstrated by Borrego et al. (2009) for the population of Portugal.

Some future work is necessary towards a fully refined, validated and transferable MEPEX and MEPEX-res model for personal exposure assessment. Some of the microenvironmental components of the model, such as the home indoor models would clearly benefit from further research into improved parameterisation. Furthermore, additional validation of the different components, as well as of the final model is considered essential.

For the future of public health research it is hoped that the development of improved methods towards personalised air pollution exposure predictions is continued, in order to better reflect individual differences in exposure, and in turn to be able to better predict and prevent related adverse health effects.

8. References

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Appendices

Appendix A: Monitoring Devices Used to Collect Particle Concentrations

Two different types of monitors have been used to collect data for PM_{2.5} and PM₁₀ for this study. Harvard Impactors have been used for the ambient monitoring campaign, Osiris/Dustmate monitors have been used for all other monitoring campaigns. In addition, concentration data from network monitoring stations, using TEOM and FDMS monitors were used in some of the analyses.

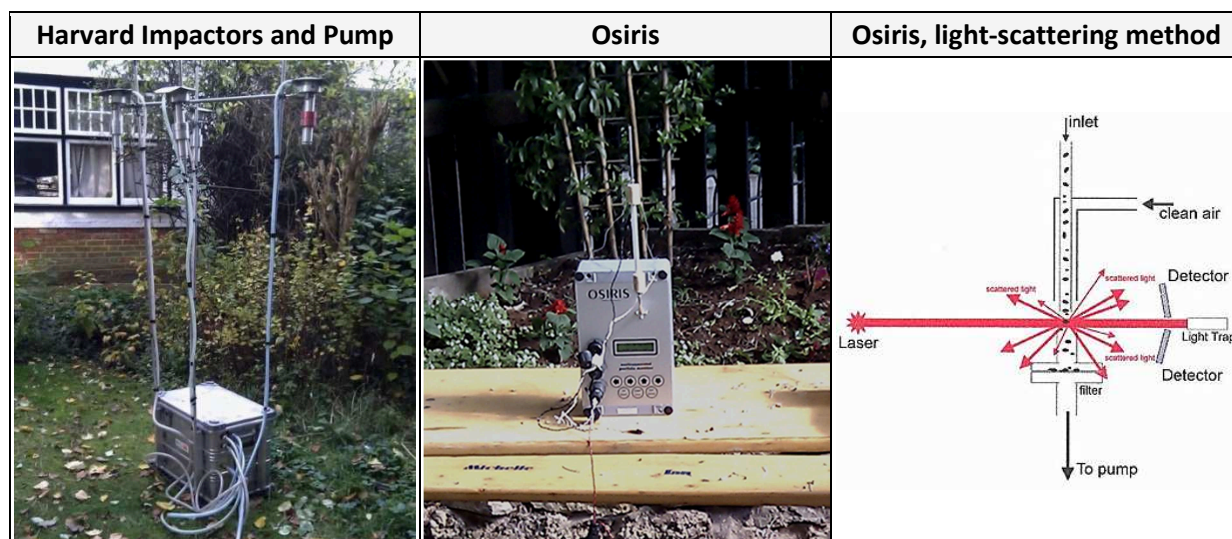
Harvard Impactor monitors

Harvard Impactors (HI) provide a method for stationary monitoring. Inside Harvard Impactors air is pumped through a filter, which accumulates particles of a certain size. The filters are later analysed in a laboratory to determine concentration gravimetrically. Other information that can be gathered from the filters includes reflectance and composition. HI have been used in this thesis to collect stationary measurements for the development of an ambient, land-use regression (LUR) model (as part of the ESCAPE study). For the use in this thesis, four Harvard Impactors were connected with one pump unit (see Appendix A, Figure 1, left). Gravimetric methods of this kind by their very design do however not provide information on short-term temporal variability, such as hours or minutes.

Osiris/Dustmate monitors

For indoor-outdoor, personal and transport monitoring devices were required to have a high temporal resolution and especially for personal and transport monitoring portability was crucial. Few types of portable monitors exist which additionally have a high temporal resolution.

Appendix A, Figure 1: Air pollution monitors used for this thesis: Harvard Impactor (left) and Osiris (middle), as well as a schematic view of the light-scattering method used by the Osiris monitor (right), source: Irving (2005).



Osiris monitors use a light scattering method to determine particle concentration at a high temporal frequency (see Appendix A, Figure 1, middle and right side). A pump draws particles into the machine through an inlet and then passes the air through a laser beam. Light scattered by the passing individual particles is then registered by a detector and converted into an electrical signal proportional to the size of the particle. A microprocessor then converts the signal to mass, assuming a material density of 1.5 grams per cubic centimetre (Irving 2005). A potential problem of the method is that water vapour could be counted as particles (Irving 2005). For all Osiris (and Dustmate) measurements in this thesis an optional heated inlet (at 60°C) was therefore used which evaporates water droplets before entering the laser inside the monitor. The effectiveness of water vapour removal was tested. The Osiris monitors (including heated inlets) were installed in a closed kitchen room. Four pots of water were heated on the stove and left to boil for more than 10 minutes, then the heat was reduced. PM concentrations remained at pre-boiling background levels.

Data was downloaded about every second day from the Osiris machine using AirQ software (by Turnkey Instruments Ltd., Northwich, UK). The Osiris monitor was sent to Turnkey Instruments Ltd. once a year for calibration according to recommendation in the user manual. Additionally, the internal clock was calibrated (to match GPS and diary recordings) every time the machine was connected to a computer.

Light-scattering devices, such as the Osiris monitors have faced some criticism about their comparability with other monitoring equipment. Both measurements using EU reference methods (or equivalent) as well as light-scattering method were however used together in the analyses for

this thesis. Concentrations recorded with the light-scattering devices were therefore calibrated to concentrations from the reference method prior to analyses.

TEOM and FDMS monitors

For some of the analyses in this thesis, concentrations from stationary measurements at existing network monitoring stations were used. TEOM and FDMS monitoring devices were used to collect data at these monitoring stations. Both monitor types collect monitoring data continuously on a sub-hourly level. TEOM and FDMS monitors both use direct mass measurements and correlate very well with concentrations monitored using reference methods (such as HI). The monitoring data from TEOM and FDMS monitors used in this study has been adjusted to EU reference equivalence by the London Air Quality Network (LAQN) or by the Department for Environment and Rural Affairs (DEFRA).

Appendix B: Comparison between Personal, Indoor and Outdoor Residential Particle Concentrations

The relationship between personal exposure and home outdoor and indoor concentrations has important implications for many studies, as studies frequently use PM concentrations at the home location as a proxy for personal exposure (see chapter 2). Indoor exposure is often directly modelled from outdoor concentrations, making the relationship between indoor and outdoor important consideration.

The datasets of all monitored (uncalibrated) personal, home indoor, and home outdoor concentrations were therefore correlated here using Spearman's rho correlation to evaluate these relationships. The comparisons are undertaken for 39 days of simultaneously collected data for Home A/ Individual A, as these provided the largest collected datasets. Spearman's rho was chosen over the more commonly applied Pearson's correlation, as it does not depend on a normal distribution of the dataset. Results for Spearman's rho correlations are presented in Appendix B, Table 1. All results are highly significant on the 0.01 level. For both PM size fractions the correlations between personal and home outdoor concentrations are low with 0.302 for PM_{2.5} and 0.117 for PM₁₀. The correlations between personal and indoor particles are in comparison higher with 0.521 for PM_{2.5} and 0.641 for PM₁₀. It should however be noted that some of the personal concentrations (when the individual is at home) use the same data as the home indoor dataset. The correlation would be lower if a second personal monitor would have been carried at the person when at home. For the comparison of home indoor to home outdoor concentrations, results for Spearman's rho are 0.440 for PM_{2.5} and 0.284 for PM₁₀.

Appendix B, Table 1: Spearman's Rho correlations between personal and home outdoor, personal and home indoor, as well as home indoor and home outdoor concentrations of PM_{2.5}, and PM₁₀ concentrations for home/ individual A, based on minute concentrations. **Correlation is significant at a 0.01 level.

	Personal – Home Outdoor	Personal – Home Indoor	Home Outdoor – Home Indoor
PM _{2.5}	0.302**	0.521**	0.440**
PM ₁₀	0.117**	0.641**	0.284**

Several other studies have looked at the relationship between personal, home outdoor, and home indoor PM_{2.5} and PM₁₀ concentrations. As described in chapter 2, several other studies have found low correlations between personal and home outdoor concentrations. Correlations between home indoor and personal concentrations have also been higher for these studies (Janssen et al. 2005;

Koistinen et al. 2001; Lai et al. 2004; Janssen et al. 1998). Spearman's rho results for home indoor to outdoor concentrations presented here lie within the range of correlation results presented in chapter 2.

In general the results presented above are similar to findings from the literature. The low correlations between outdoor home concentrations and personal exposure found in this study confirm the risk of exposure misclassification, if personal exposure is estimated from home outdoor concentrations. Higher correlations between home indoor and personal exposure may indicate that the application of a home indoor model for times at home could improve ambient exposure estimates. However, Spearman's rho correlations between outdoor and indoor home concentrations are relatively low, which limits the potential for simple indoor predictions based directly on home outdoor concentrations (i.e. I/O ratio). The comparison, above, confirms the necessity for a better understanding of different microenvironments and their influence on personal exposure, as well as the need for more refined models of personal exposure.

Appendix C: Co-locations between Osiris Monitors

Appendix C, Table 1: Spearman's Rho correlations between four co-located Osiris (and Dustmate) monitors for PM_{2.5} (in µg/m³ and based on minute data). Monitor IDs: O01, O02, O28, and D73. **Correlation is significant on the 0.01 level.

Date	Monitors O28 & D73	Monitors O28 & O02	Monitors O28 & O01	Monitors O02 & O01	Monitors O02 & D73	Monitors O01 & D73	No. of Min
22.4.2010	0.967**						314
6.5.2010	0.690**						108
18.5.2010		0.941**					807
14.6.2010		0.898**					324
25.6.2010	0.910**	-0.469**					114
28.6.2010		0.330					34
29.6.2010		0.822**					17
9.9.2010	0.892**						21
24.9.2010	0.774**	0.872**	0.820**	0.929**	0.911**	0.950**	375
2.11.2010				0.923**	0.993**	0.991**	628
10.12.2010				0.803**	0.795**	0.974**	473
14.1.2011	0.994**	0.989**	0.989**	0.990**	0.990**	0.993**	279
17.2.2011	0.428**		0.963**			0.459**	83
1.3.2011		0.962**	0.958**	0.981**			122
2.7.2011					0.493**		44
3.9.2011				0.937**			126

Appendix C, Table 2: Spearman's Rho correlations between four co-located Osiris (and Dustmate) monitors for PM₁₀ (in µg/m³ and based on minute data). Monitor IDs: O01, O02, O28, and D73. **Correlation is significant on the 0.01 level.

Date	O28 – D73	O28 – O02	O28 – O01	O02 – O01	O02 – D73	O01 – D73	No. of Min
22.4.2010	0.743**						314
6.5.2010	0.712**						108
18.5.2010		0.888**					807
14.6.2010		0.837**					324
25.6.2010	0.239**	0.505**					114
28.6.2010		0.670**					34
29.6.2010		0.789**					17
9.9.2010	0.749**						21
24.9.2010	0.554**	0.683**	0.664**	0.755**	0.623**	0.657**	375
2.11.2010				0.789**	0.878**	0.902**	628
10.12.2010				0.499**	0.509**	0.915**	473
14.1.2011	0.972**	0.952**	0.944**	0.927**	0.949**	0.955**	279
17.2.2011	0.568**		0.094			0.651**	83
1.3.2011		0.960**	0.944**	0.967**			122
2.7.2011					-0.118		44
3.9.2011				0.575**			126

Appendix D: Ambient Monitoring Campaign, Site Descriptions

Appendix D, Table 1: Site description of monitored locations for which particle concentrations were collected during the ambient monitoring campaign (as part of the ESCAPE project).

Station Number	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17	18	19	20	21
City	London	London	Oxford	Henley-o.-T.	Redding	Slough	London	London	London	London	London	London	Chadlington	London	Oxford	Maidenhead	London	London	London	London	London
Centre (C)/suburb (S)	C	S	S	S	C	S	S	S	C/S	S	S	C	Rural	C	C	S	C	C	C	S	S
Site type	Bg	Bg	Bg	Bg	Bg	Bg	Bg	Bg	Bg	Bg	Bg	Bg	Rural	Bg	Tr	Tr	Tr	Tr	Tr	Bg	Tr
Light vehicles flow on the nearest street (cars day-1):	600	2365	540	few	few	few	2189	1450	2320	few	few	few	1500	few	12354	22438	1423	17148	40232	18960	14939
Heavy vehicles flow on the nearest street (cars day-1):	few	214	few	0	0	0	93	26	20	0	0	0	50	0	543	1077	25105	4402	4207	3755	3469
Distance to nearest street (m)	50	40	40	20	25	20	15	40	20	15	25	30	25	2	2	18	22	12	3	25	8
Distance to nearest major street (m)	400	80	-	-	-	-	-	-	150	70	-	100	0	-	2	18	22	12	3	25	8
Width of the nearest street (m)	12	25	25	30	15	30	-	30	15	-	-	-	-	-	18	-	30	18	-	-	-
Height of monitored building (m)	-	5	7	5	6	6	8	6	6	10	5	6	7	-	8	7	9	10	-	6	-
Buildings uninterrupted for at least 25 meter on each side (yes / no)	no	yes	no	no	no	no	no	yes	yes	no	no	no	no	no	yes	no	no	no	no	no	no
Floor at which outdoor measurements are made	0	2	0	0	0	0	0	0	0	1	0	0	0	0	1	0	0	0	-	1	-
Sampling height for outdoor measurements (m)	1.50	2	2	2	2	2	2	2	2	4.50	2	2	2	2	4.50	3	2.50	1.50	5	4.50	3
Sampling site in backyard (B), streetside (S) or rooftop (R)	n. a.	B	B	B	B	B	B	B	B	B	B	B	B	S	S	S	S	-	S	S	S
Large parking lot within 100 meter	no	no	no	no	no	no	no	no	no	no	no	no	no	yes	no	no	no	no	no	no	no
Small industrial plant (e.g. garage, petrol station) within 100 meter?	no	no	no	no	no	no	no	no	no	no	no	no	no	no	no	no	no	50m	no	no	no

Appendix D, Figure 1: Pictures of monitored locations for which particle concentrations were collected during the ambient monitoring campaign as part of the ESCAPE project. Source of pictures 14 - 21: Google maps: (www.google.co.uk/maps, accessed between 4th Jan. 2010 and 1st May 2014).



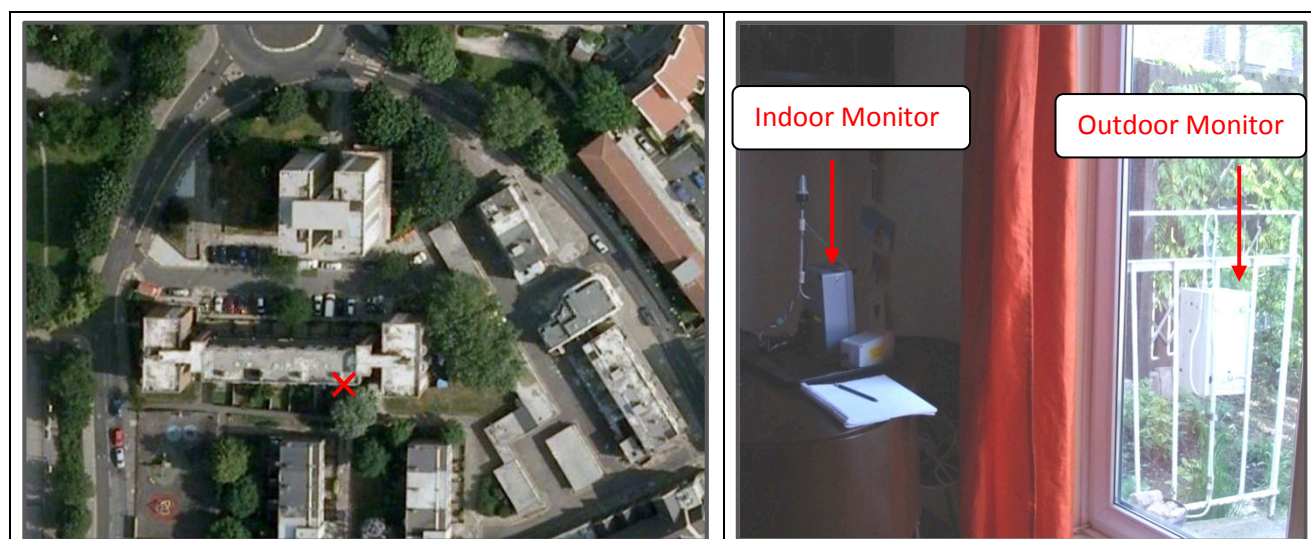


Appendix E: Description of Homes Monitored for the Indoor-Outdoor and Personal Campaigns

Home A

The majority of indoor-outdoor (as well as personal) monitoring has been collected in Home A. This home is located at a background location in a housing estate in East Central London, Shoreditch (see Figure 10 and Appendix E, Figure 1, left side). It is a Maisonette flat of 55m², with space equally distributed between two floors. Living room, hallway and kitchen are on the ground floor; bathroom and two bedrooms are on the first floor. All floors in the apartment have a smooth surface (mostly laminate). The building was built in the 1940s and is 6 storeys high. The indoor monitor was located on a table in the living room. The outdoor monitor was located in the garden, attached to a metal railing directly outside the living room and the indoor monitor (, right side). It is encased in a custom-built (Turnkey Instruments Ltd., Northwich, UK) metal box for protection. Both inlets are located at a height of 120cm and at a distance of about 70cm from the house wall and window. Two people live permanently in the apartment; both regularly leave the house during work hours. Weekday indoor and outdoor PM concentrations were recorded during 39 days.

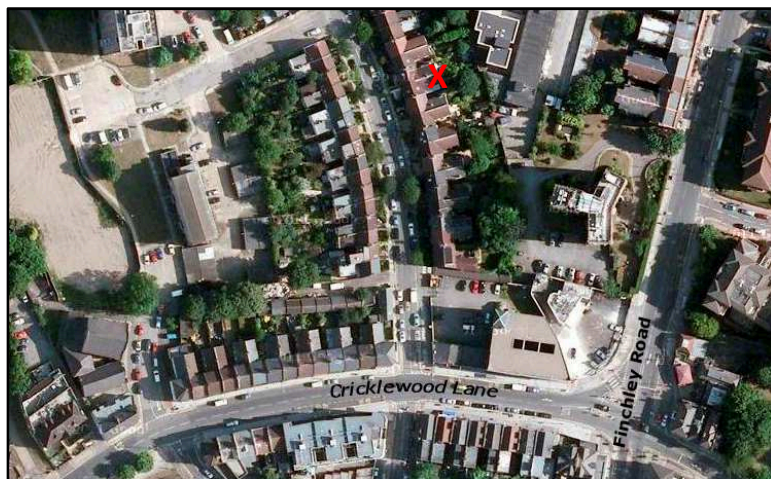
Appendix E, Figure 1: Aerial photograph of home A with home location indicated by a red cross. Source: Google maps (www.google.com/maps, accessed: 4th Octobre 2011). Right side: Indoor and outdoor monitoring location at home A.



Home B

Home B is located in Northwest London (Cricklewood) in a quiet street, but less than 100m from two major roads (Finchley Road and Cricklewood Lane) (see Figure 10 and Appendix E, Figure 2). The home is a first floor one-room apartment located in a 2-storey terraced house from the early 20th century. One occupant lives in the room, the kitchen and bathroom are however shared with six other tenants. Direct influence to the indoor air from other rooms of the house was considered unlikely as the door to the room was always closed, as were all other doors in the house (including the kitchen on the floor below). The room is 15m² (40.5 m³) in size and the floor is covered with a fitted carpet.

Appendix E, Figure 2: Aerial photograph of home B with home indicated by a red cross. Source: Google maps (www.google.com/maps, accessed: 6th Octobre 2011).



The indoor monitor was located on a small desk about 50cm from the outer wall and window, the outdoor monitor was located on the landing of a fire escape staircase about 1.50m outside the room and directly opposite the indoor monitor, both were at a high of 1m from the 1st floor level, and about 4m from the ground level. 11 days of indoor-outdoor monitoring were recorded at this home. The occupant, individual B carried a third monitor during working hours recording personal PM levels during two of the 10 days for personal monitoring.

Home C

Home C is located in a residential area of West-Central London, Pimlico (see Figure 10 and Appendix E, Figure 3, left side), a basement apartment in a five storey Victorian terraced house. The size of the

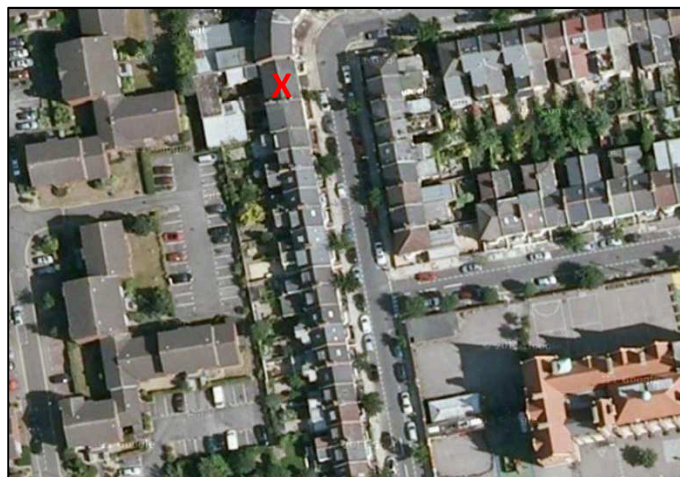
apartment is 60m² (150m³) and the floor is covered with smooth surfaces (mostly laminate). The kitchen is open plan and connected with the living room and hallway. The flat is occupied by a family of four, two adults and two children (both aged 3), who all leave the house during working hours. The indoor monitor was located in the main bedroom, on a shelf at a high of about 1.50m. The outdoor monitor was located in the inner courtyard at about 1m high, directly outside the main bedroom (see Appendix E, Figure 3, right side), which has a door opening to the courtyard. Indoor and outdoor monitoring took place during 7 consecutive days in September 2011. One adult occupant, individual C, also collected personal monitoring data by carrying a monitor with her during 2 days in June 2011.

Appendix E, Figure 3: Aerial photograph of home C with home indicated by a red cross, source: Google maps (www.google.com/maps, accessed: 4th Octobre 2011). Right side: Outdoor monitor, home C.



Home D

Appendix E, Figure 4: Aerial photograph of home D with home indicated by a red cross. Source: Google maps (www.google.com/maps, accessed: 4th Octobre 2011).



Home D is located in Shepherd's Bush, West London (see Figure 10 and) in a residential area. It is a 3-storey terraced house of 70m² (175 m³) from the second half of the 19th century, occupied by two adults. A living room and an open plan kitchen- dining room area are located on the ground floor; bathroom and bedrooms are on the first and second floors. The indoor monitor was positioned in the dining room area on a sideboard at about 1m height.

The outdoor monitor was located in the garden directly outside the back door and the dining room, also at a height of 1m. Both occupants regularly leave the house during working hours. Indoor and outdoor concentrations were recorded during eight consecutive days in July 2011 during four of the days one of the occupants, individual D, additionally carried a personal monitor with him during the day.

Appendix F: Osiris Monitor Calibration

For the preparation of a personal exposure model, monitoring data from three different types of monitors was utilised: Osiris, Harvard Impactor (HI), and TEOM/FDMS. As shown above, the different types of monitors can cause discrepancies in the results and concern has been expressed by some studies about comparability of devices using the light-scattering method (such as Osiris) in comparison to EU reference equivalent methods (such as HI or TEOM/FDMS). Braniš & Kolomazníková (2010) state that: “Collocated measurements with a reference method showed a non-linear systematic bias of the light-scattering method, limiting the use of direct concentration readings for exact exposure analysis.” J Gulliver & Briggs (2004) on the other hand report a good linear relationship ($R^2=0.83$) between concentrations monitored by OSIRIS and TEOM devices. Several studies agree that concentrations measured with Osiris monitors are at least indicative of the concentration recorded by EU reference methods (Gulliver & Briggs 2004; Walden et al. 2010). A common observation of co-location studies is however that for both $PM_{2.5}$ and PM_{10} light scattering devices, such as the OSIRIS underestimate concentrations of EU reference methods (Gulliver & Briggs 2004; De Jonge et al. 2008; Walden et al. 2010). Calibrations with a regression equation have therefore been suggested (Gulliver & Briggs 2004; Walden et al. 2010). In reference to the evidence of under-prediction of OSIRIS monitors from the literature, as well as to observations during co-locations for this thesis (see chapter 3), a decision was taken to calibrate all data measured by OSIRIS monitors.

The “Demonstration of equivalence” (EC Working Group on Guidance for the Demonstration of Equivalence 2010) is the main guideline within Europe to test for EU monitor equivalence. Several studies have been undertaken according to these guidelines, two of which include full equivalence analyses of Osiris monitors (De Jonge et al. 2008; Walden et al. 2010). One of them, a report from Finland by Walden et al. (2008), developed a regression equation that adjusts data from OSIRIS monitors to the EU reference method. The calibration equations calculated by Walden et al. (2010), were chosen, as they follow a rigorous and universally accepted protocol to establish EU reference method equivalence. Main criteria include data coverage, as well as criteria determining minimum requirements for several parameters of measurement uncertainties. The performance evaluation is undertaken for the whole dataset, as well as for data subsets (e.g. only high concentrations, certain seasons) (see also: EC Working Group on Guidance for the Demonstration of Equivalence 2010; Walden et al. 2010). For $PM_{2.5}$ however calibrated data was only accepted as indicative of EU reference measurements and only for the whole dataset, not for data subsets. For PM_{10} , full equivalence to EU reference method was achieved after calibration for the full dataset (autumn and

summer) and autumn data after calibration, but not for summer data (due to unusual concentrations in one of two monitored summer months) (Walden et al. 2010). The calibration equations for PM_{2.5} and PM₁₀ by Walden et al. (2010) can be written as follows:

Equation 17 for PM_{2.5}:

$$y_{\text{REF}} = -0.11 + 1.79 \cdot x_{\text{OSIRIS}} \quad (\text{also: } x_{\text{OSIRIS}} = 0.06 + 0.56 \cdot y_{\text{REF}})$$

Equation 18 for PM₁₀:

$$y_{\text{REF}} = -1.10 + 1.42 \cdot x_{\text{OSIRIS}} \quad (\text{also: } x_{\text{OSIRIS}} = 0.78 + 0.70 \cdot y_{\text{REF}})$$

With y_{REF} , as the adjusted particle concentrations, and x_{OSIRIS} , as the original concentrations collected using Osiris monitors.

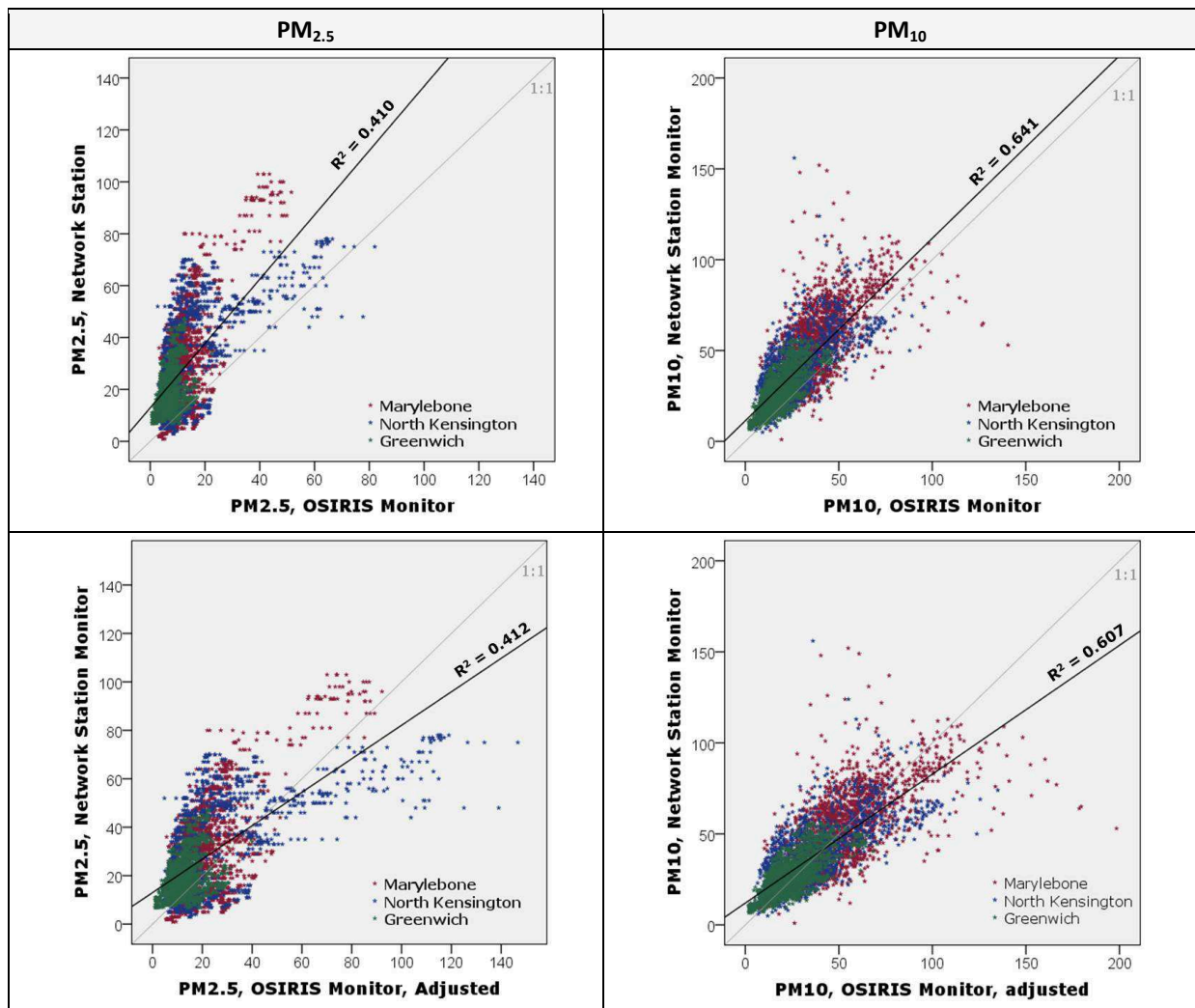
Monitor co-location in London

Concentrations were collected for Osiris monitors in co-location with TEOM/FDMS monitors, as described in chapter 3. The resulting data is used here to compare the relationship between Osiris and FDMS/TEOM monitors before and after calibration. The data is analysed at a 15 minute level, the highest temporal resolution available from the LAQN for concentrations monitored by FDMS/TEOM. The three locations have been pooled in order to reflect a variety of locations. Only weekdays were included as this PHD focuses on predicting weekday exposure and weekends may have been influenced by different factors and for example do not include typical rush hour traffic.

The data is presented in Appendix F, Figure 1 before (upper part) and after (lower part) calibration with Equation 17 and Equation 18 for PM_{2.5} (left side) and PM₁₀ (right side) respectively. The scatter plots before calibration show for PM_{2.5} an R^2 result of 0.410 with clear underprediction of concentrations monitored by the Osiris equipment for most data points. The R^2 result for PM₁₀ is higher with 0.641. Much of the data however lies above the 1:1 reference line, showing underprediction of the concentrations recorded by the Osiris monitor. Calibrations to EU equivalence only slightly changed R^2 results to 0.412 for PM_{2.5} and to 0.607 for PM₁₀. For both particle size fractions the data points show however a clear shift towards the reference line compared to pre-calibration results. In general, the results of the co-location show only medium high R^2 results between the two types of monitors. Not all of the observed differences between concentrations can be attributed to differences between equipment. Particle concentrations may vary over very short distances. Locations of the monitor inlets were for all co-locations a few meters apart, which may have resulted in additional differences between concentrations recorded by the monitoring devices. Adjusting concentration recorded by the Osiris monitors to EU reference equivalent, clearly reduces underprediction for much of the dataset, both for PM_{2.5} and PM₁₀. It

does however not improve correlation between concentrations monitored by different devices and a few data points overpredict after calibrations.

Appendix F, Figure 1: OSIRIS monitoring concentrations against concentrations at network monitoring stations of $PM_{2.5}$ and PM_{10} (both in $\mu g/m^3$) for co-located monitors before (upper part) and after (lower part) calibration to EU reference equivalence.



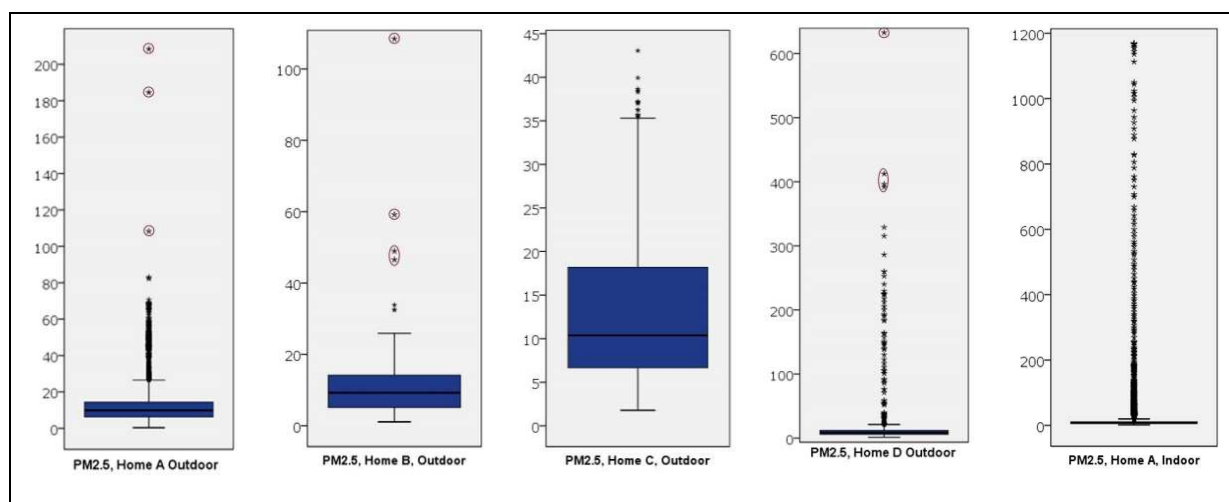
For the analyses undertaken as part of this thesis (chapter 5 and 6), adjusted OSIRIS concentrations of $PM_{2.5}$ need to be viewed with caution for two reasons: 1. The calibration equation only adjusts concentrations to be indicative of EU reference methods, and 2. The concentrations collected by Osiris monitors as part of this thesis have shown R^2 results of below 0.5 when compared to an EU reference equivalent method. The equation for the calibration of PM_{10} has shown full reference equivalence by Walden et al. (2010). Calibrated PM_{10} concentrations can therefore be compared and discussed with confidence and are generally regarded as EU reference equivalent. For a few single minute data points (of all datasets monitored by Osiris) the adjusted concentrations resulted in

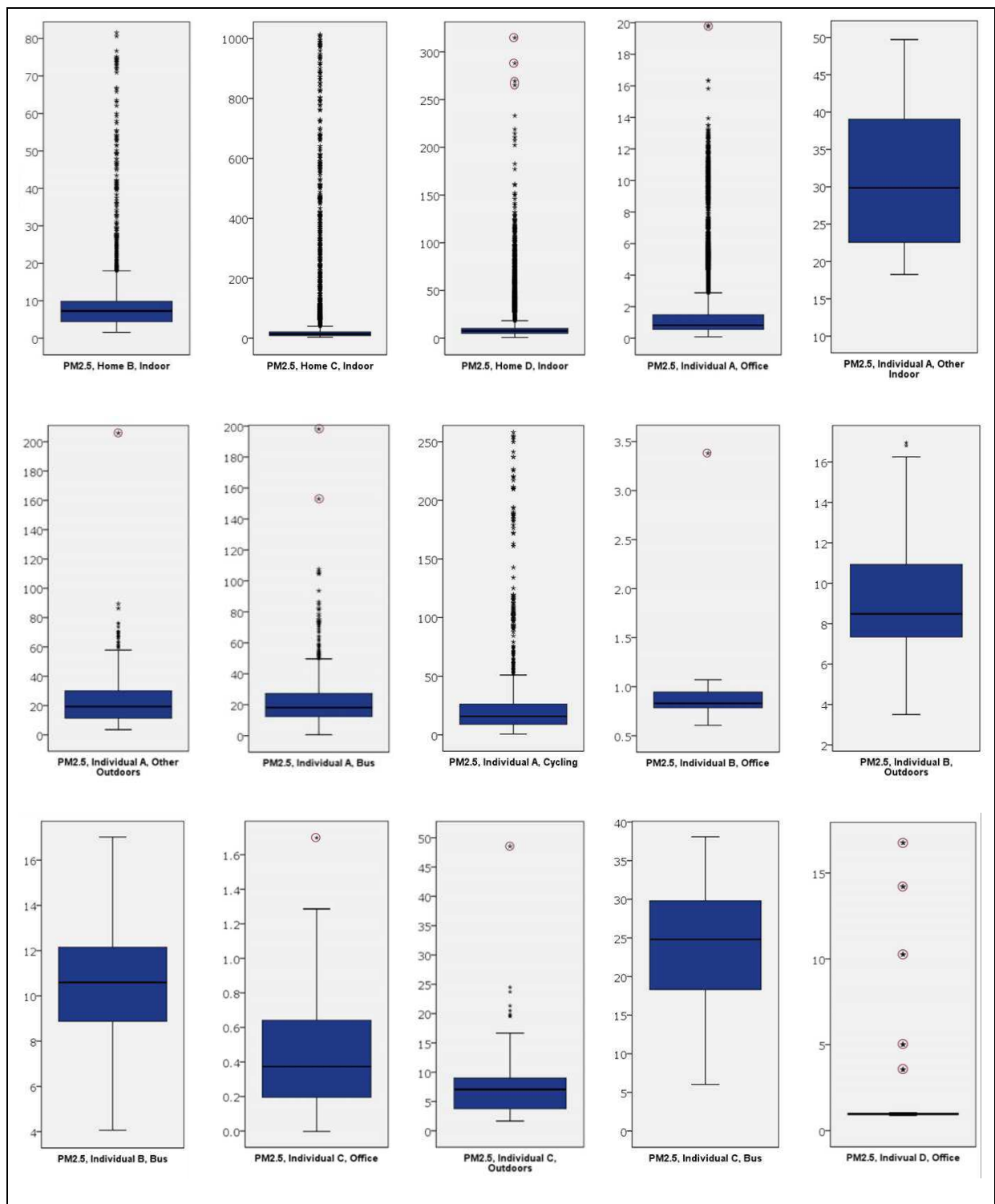
negative values, these were consequently deleted. All distributions of the calibrated data by dataset and ME are presented in Appendix G.

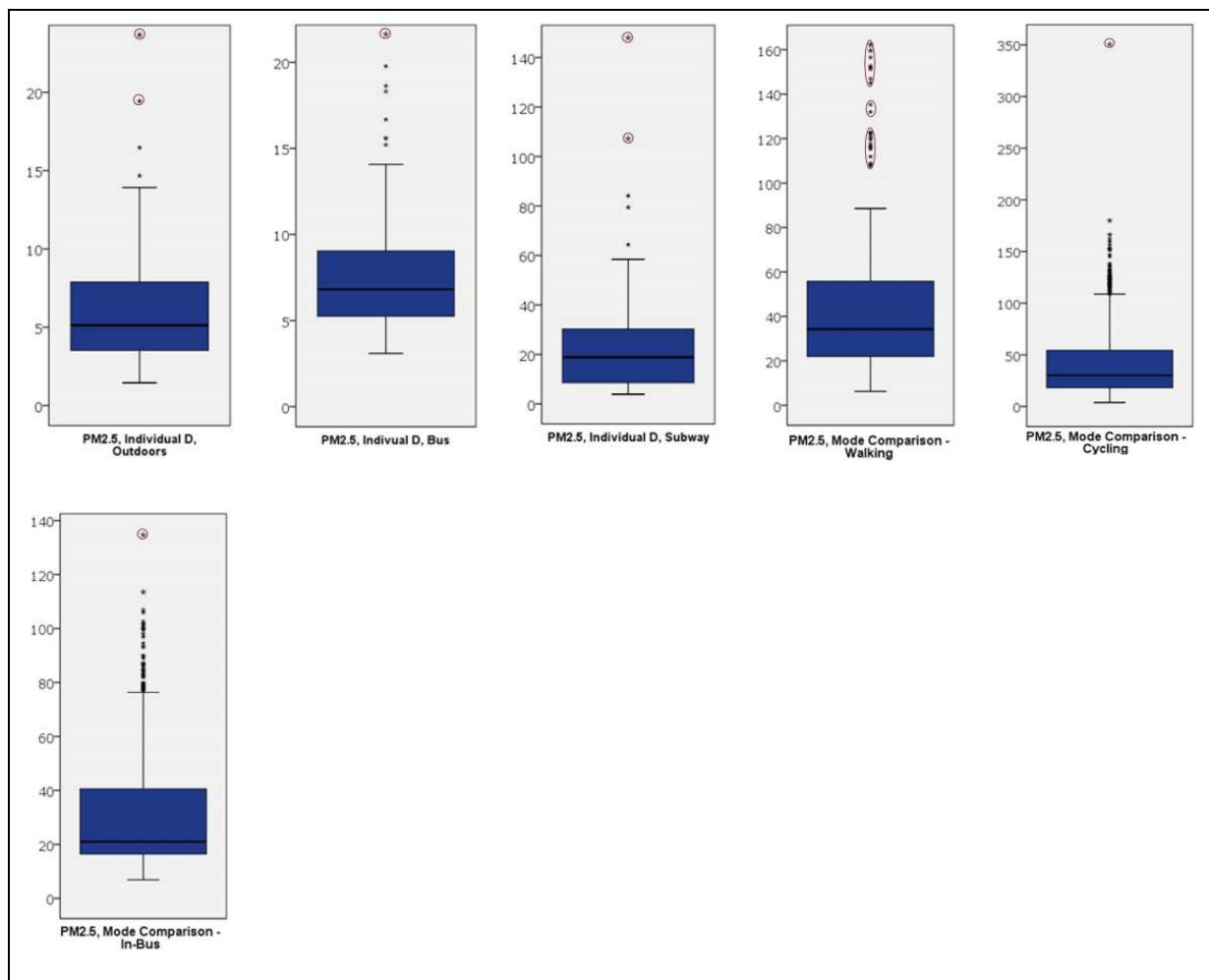
Appendix G: Distribution of Monitoring Data Collected using Osiris Monitors, after Calibration

In the following figures (Appendix G, Figure 1Error! Reference source not found. and Appendix G, Figure 2Error! Reference source not found.) the data distributions of the calibrated concentrations for PM_{2.5} and PM₁₀ minute concentrations are presented. Presentation of data collected during the personal exposure monitoring campaign is undertaken for each microenvironment separately. This decision was taken in reference to the fact that there are substantial differences in the data distribution of different microenvironments. As presented in chapter 3, the distribution of particle concentrations in a bus is for example different compared to the concentration distribution in the office microenvironment. Unusual values have for this reason also been identified separately by microenvironment (unusual values are marked in red). The more commonly applied removal of unusual values for the entire datasets would have resulted in the removal of only few (mainly cooking related) concentrations. The identification of extreme values has furthermore been described in chapter 3.

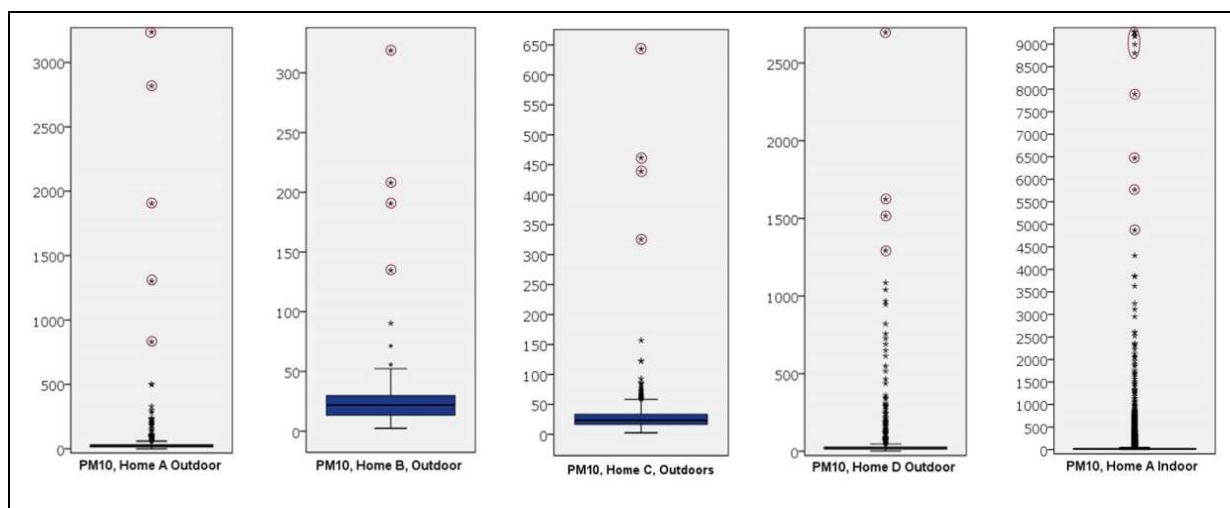
Appendix G, Figure 1: Distributions of data collected during the indoor, outdoor, and personal and transport monitoring campaigns for calibrated PM_{2.5} concentrations by campaign and microenvironment. Unusual values are marked in red.

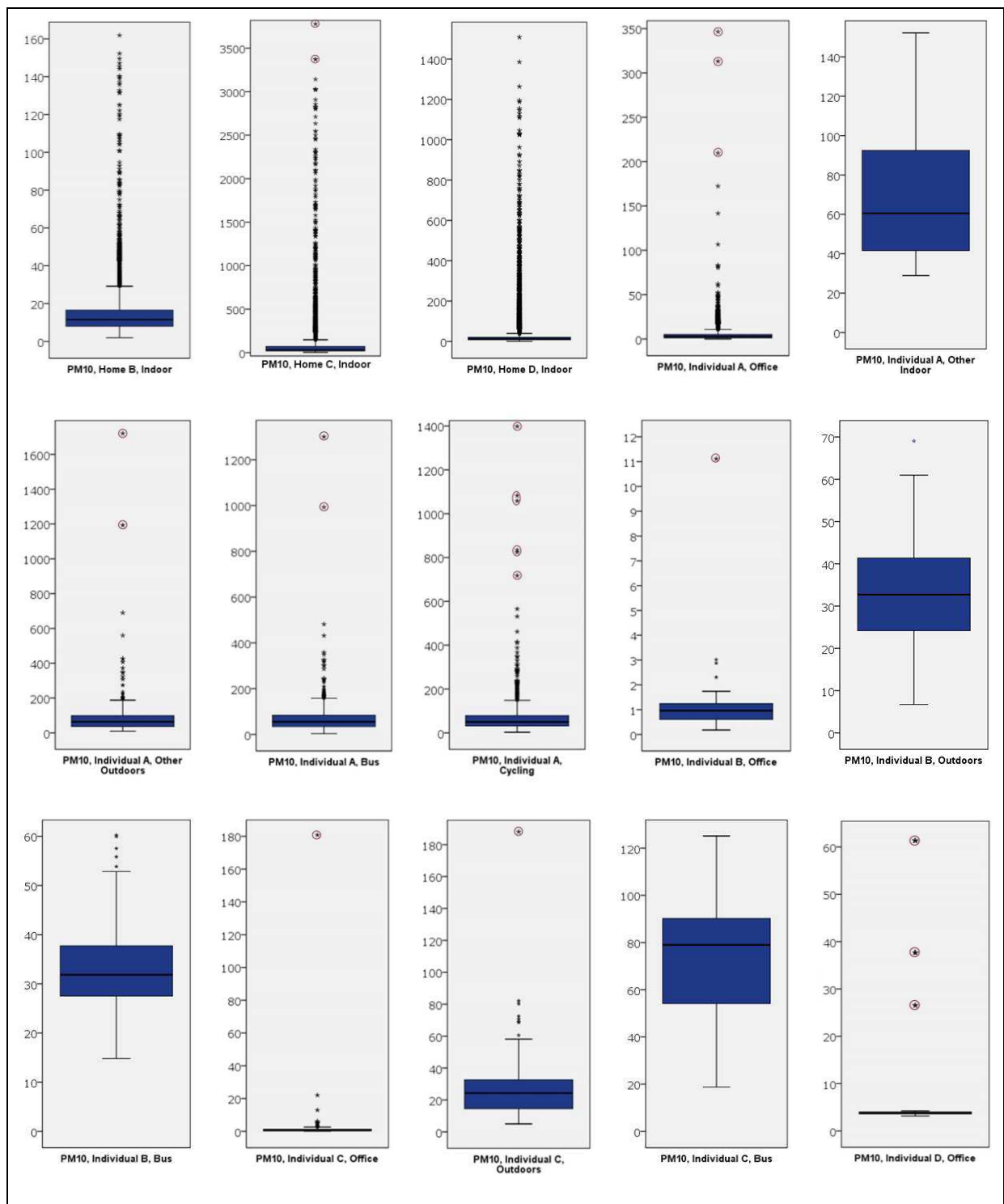


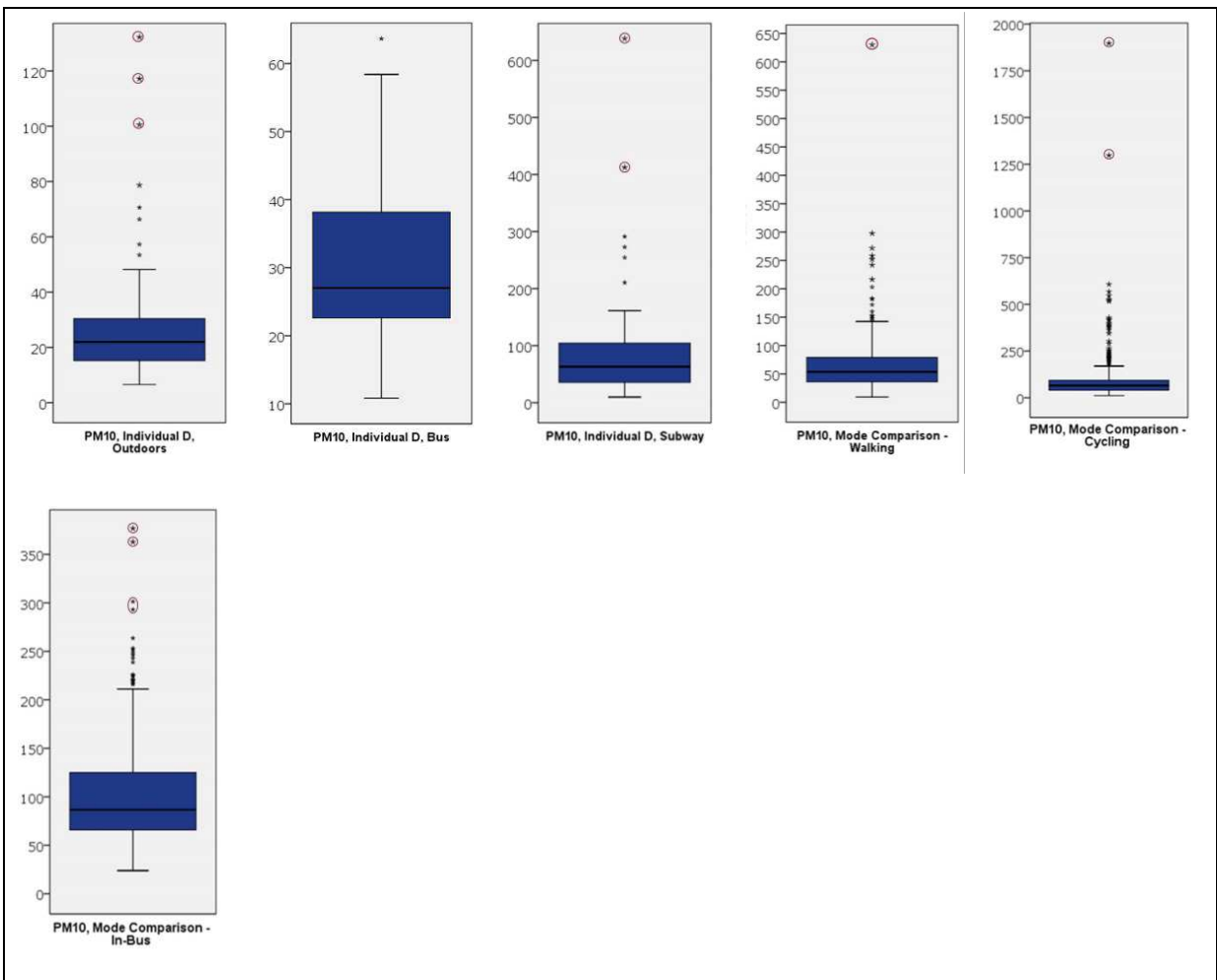




Appendix G, Figure 2: Distributions of data collected during the indoor, outdoor, and personal and transport monitoring campaigns for calibrated PM10 concentrations by campaign and microenvironment. Unusual values are marked in red.







Appendix H: Additional Information on Temporal Adjustments of the LUR Model

Appendix H1: Descriptive Statistics for Datasets Used in the Development of a Temporal Adjustment of the LUR Model

Appendix H, Table 1: Descriptive statistics (Mean, SD, Range) for 2010 data of daily and weekly PM_{2.5} (in µg/m³) concentrations for London monitoring stations (Station ID given in chapter 4).

	Station ID	Daily				Weekly			
		n	Mean	SD	Range	n	Mean	SD	Range
Roadside	1	309	14.27	5.25	30.46	45	14.25	2.50	10.15
	2	324	11.38	5.00	34.38	48	11.59	3.47	13.30
	3	365	15.10	9.09	66.08	53	15.48	6.48	31.63
	4	357	16.97	10.09	72.38	53	17.22	6.46	30.11
	5	361	16.40	6.01	53.33	53	16.55	3.13	13.29
	6	339	20.22	8.87	57.78	52	20.48	6.56	38.04
	7	356	13.48	4.61	32.42	53	13.54	2.34	10.87
	8	365	17.51	9.21	54.78	53	17.80	6.21	25.98
	9	355	18.17	8.89	50.58	53	18.44	5.76	30.08
Background	10	331	16.20	8.86	70.82	50	16.29	6.08	24.85
	11	346	12.75	7.35	48.26	53	12.92	4.71	17.68
	12	345	13.95	7.21	39.88	52	14.19	4.68	18.61
	13	281	13.46	7.80	50.49	43	13.75	5.35	19.38

Appendix H, Table 2: Descriptive statistics (Mean, SD, Range) for 2010 data of daily and weekly PM₁₀ (in µg/m³) concentrations for London monitoring stations (Station ID given in chapter 4).

	Station ID	Daily				Weekly			
		n	Mean	SD	Range	n	Mean	SD	Range
Roadside	1	353	27.99	9.83	55.97	53	28.42	6.24	30.66
	2	306	32.82	13.42	120.75	45	33.25	8.56	44.34
	3	323	25.76	9.08	52.06	47	25.78	5.38	25.88
	4	326	26.12	10.63	59.03	49	26.45	6.82	25.28
	5	320	29.49	8.22	43.25	49	29.61	5.00	21.73
	6	364	37.51	13.18	79.43	53	37.54	6.45	28.47

	Station ID	Daily				Weekly			
		n	Mean	SD	Range	n	Mean	SD	Range
	7	315	23.96	9.02	52.00	47	24.39	6.31	28.94
	8	352	28.07	11.42	63.86	53	28.27	6.80	25.21
	9	365	21.89	8.72	59.13	53	22.14	5.75	25.96
	10	365	28.28	11.10	76.88	53	28.54	6.52	28.88
	11	365	32.58	10.94	66.08	53	32.71	5.50	26.12
	12	358	23.20	17.72	272.13	53	23.68	10.59	70.51
	13	324	23.08	10.02	54.71	49	23.16	6.19	22.13
	14	347	28.27	9.36	47.30	53	28.21	5.93	26.27
	15	364	27.02	8.70	50.00	53	27.18	4.94	22.02
	16	295	32.92	9.65	76.33	47	32.60	5.30	29.68
	17	316	19.72	8.17	66.83	48	19.76	5.33	25.55
	18	361	25.02	9.18	65.33	53	25.23	5.90	28.33
	19	285	26.78	15.55	183.00	45	26.69	7.94	35.80
	19b	285	25.61	10.30	64.33	45	25.63	6.23	24.42
	20	358	23.09	8.91	51.71	53	23.19	5.09	21.83
	21	288	30.84	10.46	61.83	46	30.31	5.80	23.79
	22	361	20.81	8.26	53.17	53	21.03	5.21	22.63
	23	341	24.55	9.85	53.03	51	24.85	5.46	25.04
	24	333	29.06	8.29	53.24	51	29.55	6.69	41.14
	25	365	29.76	9.98	67.04	53	30.01	5.95	25.63
	26	359	35.38	12.47	60.10	53	35.37	6.64	29.43
Background	27	351	20.00	8.27	47.71	52	20.21	5.34	20.57
	28	333	17.81	8.25	51.45	51	18.02	6.28	27.98
	29	354	19.88	8.51	48.88	51	19.88	5.31	21.94
	30	325	26.39	10.60	93.93	50	26.26	5.69	27.74
	31	362	16.94	7.73	54.13	53	17.04	4.44	22.15
	32	322	21.21	9.53	69.97	51	21.26	6.06	29.17
	33	365	21.79	8.45	50.13	53	22.02	5.23	24.35
	34	359	20.29	8.60	50.91	53	20.39	5.50	20.68
	35	352	21.67	8.96	52.33	52	21.87	5.54	23.06
	36	357	21.80	8.12	55.25	52	22.05	4.98	21.05

Appendix H, Table 3: Descriptive statistics (Mean, SD, Range) for 2011 data of hourly, daily, and weekly PM_{2.5} (in µg/m³) concentrations for London monitoring stations (Station ID given in chapter 4).

Station ID	Hourly				Daily				Weekly			
	n	Mean	SD	Range	n	Mean	SD	Range	n	Mean	SD	Range
1	7592	15.66	8.61	69	323	15.72	6.62	36.96	47	15.57	4.58	19.88

	Hourly				Daily				Weekly			
Station ID	n	Mean	SD	Range	n	Mean	SD	Range	n	Mean	SD	Range
3	8257	18.70	14.61	112	350	18.69	12.82	75.38	53	18.30	9.61	44.30
5	8724	17.26	8.15	74	365	17.26	5.63	33.58	53	17.14	3.47	16.16
7	8219	15.35	7.02	76	355	15.35	5.25	31.25	53	15.23	3.54	16.00
9	8270	17.62	14.60	142	358	17.50	12.49	76.79	53	17.27	9.55	49.08

Appendix H, Table 4: Descriptive statistics (Mean, SD, Range) for 2011 data of hourly, daily, and weekly PM₁₀ (in µg/m³) concentrations for London monitoring stations (Station ID given in chapter 4).

	Hourly				Daily				Weekly			
Station ID	n	Mean	SD	Range	n	Mean	SD	Range	n	Mean	SD	Range
2	7624	34.51	20.36	233	322	34.57	16.54	92.33	47	34.17	13.12	55.74
3	7680	25.48	13.90	113	328	25.54	11.63	68.19	49	25.33	9.19	45.19
4	8224	26.82	17.35	123	357	26.67	14.57	82.79	53	26.59	11.83	52.02
5	8063	31.57	16.41	283	344	31.68	12.50	86.69	51	31.27	9.44	55.62
6	8675	36.56	21.01	511	365	36.59	14.09	84.71	53	36.22	10.11	55.69
7	8522	25.09	14.97	116	361	25.06	12.84	82.29	53	24.86	10.11	54.01
8	8213	26.29	18.75	511	352	26.34	13.69	82.67	52	26.37	10.28	47.75
9	8724	23.20	14.72	182	365	23.20	12.76	85.29	53	22.98	9.98	52.68
10	8710	32.07	17.61	172	365	32.19	15.13	121.87	53	31.86	11.62	66.83
11	8728	34.70	20.03	462	365	34.70	13.66	89.29	53	34.38	9.59	50.42
12	8555	23.29	17.70	204	361	23.39	15.26	100.33	53	23.28	12.44	60.92
14	8248	31.38	16.82	304	351	31.40	13.48	96.50	53	30.94	10.53	59.51
15	8583	29.28	15.95	510	361	29.29	12.25	75.04	53	28.96	9.31	47.35
16	6948	37.00	19.39	531	313	37.15	15.03	123.68	49	36.70	10.68	58.40
18	8313	25.80	14.17	139	357	25.81	11.85	74.36	53	25.59	8.84	45.75
20	8569	27.54	14.88	153	361	27.52	11.91	66.96	53	27.25	9.59	41.21
21	6858	25.95	14.23	143	301	26.00	11.75	63.37	47	25.84	9.63	36.67
22	8693	23.21	13.62	120	365	23.20	11.78	77.00	53	22.96	9.17	48.42
24	8687	31.98	14.37	145	365	32.02	11.15	78.67	53	31.78	8.28	46.75
26	8542	40.65	18.79	419	364	40.69	13.84	82.08	53	40.33	10.25	50.68

Appendix H, Table 5: Descriptive statistics (Mean, SD, Range) of hourly, daily, and weekly PM_{2.5} (in µg/m³) concentrations of 2010 for monitoring stations outside London. Station type is added as: UB = urban background, RB = rural background.

PM _{2.5}		Hourly				Daily				Weekly			
Station	Type	n	Mean	SD	Range	n	Mean	SD	Range	n	Mean	SD	Range
Reading New Town	UB	7112	13.37	9.44	99	302	13.38	7.60	45.43	13.53	5.86	21.97	13.53
Norwich Lakenfields	UB	8170	12.69	11.00	220	345	12.67	8.51	44.96	12.89	5.98	26.10	12.89
Harwell	RB	8634	10.28	7.89	67	365	10.26	6.56	39.88	10.41	4.69	21.19	10.41

PM _{2.5}		Hourly				Daily				Weekly			
Station	Type	n	Mean	SD	Range	n	Mean	SD	Range	n	Mean	SD	Range
Bristol St Paul's	UB	8396	14.21	13.86	321	358	14.19	9.91	97.04	14.26	6.39	22.24	14.26
Leeds Centre	UB	8646	14.94	11.00	220	364	14.95	8.68	59.96	14.94	5.42	21.04	14.94
Newcastle Centre	UB	7998	10.39	8.012	85	338	10.37	6.45	40.61	10.26	4.12	15.37	10.26
Nottingham Centre	UB	6691	15.86	13.32	364	289	16.03	9.78	59.31	16.39	6.93	35.01	16.39
Oxford St Ebbes	UB	8421	14.34	9.211	98	360	14.37	7.61	43.07	14.54	5.55	21.34	14.54

Appendix H, Table 6: Descriptive statistics (Mean, SD, and Range) of hourly, daily, and weekly PM₁₀ (in µg/m³) concentrations of 2010 for monitoring stations outside London. Station type is added as: UB = urban background, RB = rural background, RS = roads

Station	Type	Hourly				Daily				Weekly			
		n	Mean	SD	Range	n	Mean	SD	Range	n	Mean	SD	Range
Norwich Lakenfields	UB	6795	17.80	11.25	129	291	17.68	8.74	47.92	45	17.60	5.90	27.03
Thurrock Stanford-le-Hope	RS	7791	20.66	12.19	221	337	20.78	9.18	55.71	52	21.33	5.84	27.19
Mole Valley Dorking	UB	8397	18.98	10.00	194	357	18.93	7.48	39.67	53	19.04	5.46	29.88
Harwell	RB	6701	15.99	10.10	122	291	16.03	8.22	49.50	46	16.15	6.50	32.46
Bristol St Paul's	UB	8442	20.33	15.24	480	359	20.30	10.41	110.38	53	20.22	6.42	25.81
Leeds Centre	UB	8607	20.97	13.29	237	364	21.05	9.89	67.92	53	20.95	6.18	22.60
Newcastle Centre	UB	8534	14.85	8.725	91	361	14.82	6.96	43.48	53	14.82	4.61	23.72
Nottingham Centre	UB	7788	23.00	18.40	851	334	23.02	11.16	70.71	51	23.53	8.57	42.10
Oxford St Ebbes	UB	8073	16.67	9.919	133	345	16.81	8.01	46.00	52	16.81	5.71	24.04

Appendix H2: Ratios for Adjustment B (using temporal patterns) of the LUR Model

Appendix H, Table 7: Seasonal, daily, and hourly ratios used to calculate temporal adjustments for a land-use regression model using temporal patterns (adjustment version B) for PM_{2.5}.

PM _{2.5}	Spring							Summer							Autumn							Winter									
Season:	0.98							0.74							0.98							1.31									
	Mon	Tue	Wed	Thu	Fri	Sat	Sun		Mon	Tue	Wed	Thu	Fri	Sat	Sun		Mon	Tue	Wed	Thu	Fri	Sat	Sun		Mon	Tue	Wed	Thu	Fri	Sat	Sun
Daily:	1.01	1.01	1.04	0.96	0.94	0.94	0.95		0.76	0.76	0.78	0.72	0.71	0.71	0.71		1.01	1.01	1.05	0.97	0.94	0.95	0.95		1.35	1.35	1.39	1.29	1.26	1.27	1.27
Hourly:																															
0	1.03	1.03	1.06	0.98	0.96	0.96	0.97		0.77	0.77	0.80	0.74	0.72	0.72	0.73		1.03	1.03	1.07	0.98	0.96	0.97	0.97		1.37	1.38	1.42	1.31	1.28	1.29	1.29
1	1.00	1.00	1.03	0.95	0.93	0.94	0.94		0.75	0.75	0.77	0.72	0.70	0.70	0.71		1.00	1.00	1.04	0.96	0.93	0.94	0.94		1.34	1.34	1.38	1.27	1.24	1.25	1.26
2	0.98	0.98	1.01	0.94	0.91	0.92	0.92		0.74	0.74	0.76	0.70	0.69	0.69	0.69		0.99	0.99	1.02	0.94	0.92	0.93	0.93		1.31	1.31	1.36	1.25	1.22	1.23	1.24
3	0.97	0.97	1.00	0.92	0.90	0.91	0.91		0.73	0.73	0.75	0.70	0.68	0.68	0.69		0.98	0.98	1.01	0.93	0.91	0.91	0.92		1.30	1.30	1.34	1.24	1.21	1.22	1.22
4	0.96	0.96	0.99	0.92	0.90	0.90	0.91		0.72	0.72	0.75	0.69	0.67	0.68	0.68		0.97	0.97	1.00	0.92	0.90	0.91	0.91		1.29	1.29	1.33	1.23	1.20	1.21	1.21
5	0.99	0.99	1.02	0.94	0.92	0.93	0.93		0.74	0.74	0.77	0.71	0.69	0.70	0.70		0.99	0.99	1.03	0.95	0.93	0.93	0.94		1.32	1.32	1.37	1.26	1.23	1.24	1.25
6	1.03	1.03	1.07	0.98	0.96	0.97	0.97		0.78	0.78	0.80	0.74	0.72	0.73	0.73		1.04	1.04	1.07	0.99	0.97	0.97	0.98		1.38	1.38	1.43	1.32	1.29	1.30	1.30
7	1.06	1.06	1.10	1.01	0.99	1.00	1.00		0.80	0.80	0.83	0.76	0.75	0.75	0.75		1.07	1.07	1.10	1.02	1.00	1.00	1.01		1.42	1.43	1.47	1.36	1.33	1.34	1.34
8	1.08	1.08	1.12	1.03	1.01	1.02	1.02		0.82	0.82	0.84	0.78	0.76	0.76	0.77		1.09	1.09	1.13	1.04	1.02	1.02	1.03		1.45	1.45	1.50	1.38	1.35	1.36	1.37
9	1.05	1.05	1.08	1.00	0.97	0.98	0.98		0.79	0.79	0.81	0.75	0.73	0.74	0.74		1.05	1.05	1.09	1.00	0.98	0.99	0.99		1.40	1.40	1.45	1.34	1.31	1.32	1.32
10	1.00	1.00	1.03	0.96	0.93	0.94	0.94		0.75	0.75	0.78	0.72	0.70	0.71	0.71		1.01	1.01	1.04	0.96	0.94	0.95	0.95		1.34	1.34	1.39	1.28	1.25	1.26	1.26
11	0.97	0.97	1.00	0.93	0.91	0.91	0.91		0.73	0.73	0.75	0.70	0.68	0.69	0.69		0.98	0.98	1.01	0.93	0.91	0.92	0.92		1.30	1.30	1.34	1.24	1.21	1.22	1.23
12	0.95	0.95	0.98	0.90	0.88	0.89	0.89		0.71	0.71	0.73	0.68	0.66	0.67	0.67		0.95	0.95	0.98	0.91	0.89	0.89	0.89		1.27	1.27	1.31	1.21	1.18	1.19	1.19
13	0.93	0.93	0.95	0.88	0.86	0.87	0.87		0.70	0.70	0.72	0.66	0.65	0.65	0.65		0.93	0.93	0.96	0.89	0.87	0.87	0.88		1.24	1.24	1.28	1.18	1.15	1.16	1.17
14	0.90	0.91	0.93	0.86	0.84	0.85	0.85		0.68	0.68	0.70	0.65	0.63	0.64	0.64		0.91	0.91	0.94	0.87	0.85	0.85	0.86		1.21	1.21	1.25	1.16	1.13	1.14	1.14
15	0.91	0.91	0.94	0.87	0.85	0.85	0.86		0.68	0.68	0.71	0.65	0.64	0.64	0.64		0.91	0.92	0.94	0.87	0.85	0.86	0.86		1.22	1.22	1.26	1.16	1.14	1.14	1.15
16	0.93	0.93	0.96	0.89	0.87	0.88	0.88		0.70	0.70	0.73	0.67	0.65	0.66	0.66		0.94	0.94	0.97	0.90	0.87	0.88	0.88		1.25	1.25	1.29	1.19	1.16	1.17	1.18
17	0.96	0.96	0.99	0.92	0.90	0.90	0.91		0.72	0.72	0.75	0.69	0.67	0.68	0.68		0.97	0.97	1.00	0.92	0.90	0.91	0.91		1.29	1.29	1.33	1.23	1.20	1.21	1.21
18	1.00	1.00	1.03	0.95	0.93	0.93	0.94		0.75	0.75	0.77	0.72	0.70	0.70	0.71		1.00	1.00	1.03	0.96	0.93	0.94	0.94		1.34	1.34	1.38	1.27	1.24	1.25	1.26
19	1.05	1.05	1.09	1.01	0.98	0.99	0.99		0.79	0.79	0.82	0.76	0.74	0.74	0.75		1.06	1.06	1.09	1.01	0.99	0.99	1.00		1.41	1.41	1.46	1.35	1.32	1.33	1.33
20	1.10	1.10	1.13	1.05	1.02	1.03	1.03		0.83	0.83	0.85	0.79	0.77	0.78	0.78		1.11	1.11	1.14	1.05	1.03	1.04	1.04		1.47	1.47	1.52	1.40	1.37	1.38	1.39
21	1.13	1.13	1.16	1.07	1.05	1.06	1.06		0.85	0.85	0.88	0.81	0.79	0.80	0.80		1.13	1.13	1.17	1.08	1.06	1.06	1.07		1.51	1.51	1.56	1.44	1.41	1.42	1.42
22	1.12	1.12	1.15	1.06	1.04	1.05	1.05		0.84	0.84	0.87	0.80	0.78	0.79	0.79		1.12	1.12	1.16	1.07	1.05	1.05	1.06		1.50	1.50	1.54	1.43	1.39	1.40	1.41
23	1.06	1.06	1.10	1.02	0.99	1.00	1.00		0.80	0.80	0.83	0.76	0.75	0.75	0.75		1.07	1.07	1.11	1.02	1.00	1.00	1.01		1.43	1.43	1.47	1.36	1.33	1.34	1.34

Appendix H, Table 8: Seasonal, daily, and hourly ratios used to calculate temporal adjustments for a land-use regression model using temporal patterns (adjustment version B) for PM₁₀.

PM ₁₀	Spring							Summer							Autumn							Winter									
Season:	0.98							0.94							1.02							1.06									
	Mon	Tue	Wed	Thu	Fri	Sat	Sun		Mon	Tue	Wed	Thu	Fri	Sat	Sun		Mon	Tue	Wed	Thu	Fri	Sat	Sun		Mon	Tue	Wed	Thu	Fri	Sat	Sun
Daily:	0.99	1.02	1.04	0.98	0.98	0.93	0.91		0.95	0.97	0.99	0.95	0.94	0.89	0.87		1.04	1.06	1.07	1.02	1.03	0.98	0.95		1.08	1.10	1.12	1.07	1.07	1.01	0.99
Hourly:																															
0	0.95	0.98	1.00	0.94	0.94	0.89	0.87		0.91	0.93	0.95	0.91	0.90	0.86	0.83		0.99	1.02	1.03	0.98	0.99	0.94	0.91		1.03	1.06	1.08	1.03	1.02	0.97	0.94
1	0.91	0.94	0.95	0.90	0.90	0.85	0.83		0.87	0.89	0.91	0.87	0.86	0.82	0.80		0.95	0.97	0.98	0.94	0.94	0.89	0.87		0.98	1.01	1.03	0.98	0.98	0.93	0.90
2	0.88	0.91	0.92	0.87	0.87	0.82	0.80		0.84	0.86	0.88	0.84	0.83	0.79	0.77		0.92	0.94	0.95	0.91	0.91	0.86	0.84		0.95	0.98	1.00	0.95	0.95	0.90	0.87
3	0.87	0.90	0.91	0.86	0.86	0.82	0.79		0.83	0.85	0.87	0.83	0.83	0.78	0.76		0.91	0.93	0.94	0.90	0.90	0.86	0.83		0.94	0.97	0.99	0.94	0.94	0.89	0.86
4	0.85	0.88	0.89	0.85	0.84	0.80	0.78		0.81	0.84	0.85	0.81	0.81	0.77	0.75		0.89	0.91	0.92	0.88	0.88	0.84	0.82		0.92	0.95	0.96	0.92	0.92	0.87	0.85
5	0.87	0.90	0.91	0.86	0.86	0.81	0.79		0.83	0.85	0.87	0.83	0.82	0.78	0.76		0.91	0.93	0.94	0.90	0.90	0.85	0.83		0.94	0.97	0.98	0.94	0.93	0.89	0.86
6	0.94	0.96	0.98	0.93	0.92	0.88	0.85		0.89	0.92	0.93	0.89	0.89	0.84	0.82		0.98	1.00	1.01	0.96	0.97	0.92	0.89		1.01	1.04	1.06	1.01	1.01	0.95	0.93
7	1.01	1.04	1.06	1.00	1.00	0.95	0.92		0.97	0.99	1.01	0.96	0.96	0.91	0.89		1.06	1.08	1.09	1.04	1.05	0.99	0.97		1.09	1.12	1.14	1.09	1.09	1.03	1.00
8	1.08	1.12	1.14	1.07	1.07	1.01	0.99		1.03	1.06	1.08	1.03	1.03	0.97	0.95		1.13	1.16	1.17	1.12	1.12	1.06	1.03		1.17	1.20	1.22	1.17	1.16	1.10	1.07
9	1.11	1.14	1.16	1.10	1.09	1.04	1.01		1.06	1.08	1.10	1.05	1.05	0.99	0.97		1.15	1.18	1.19	1.14	1.14	1.09	1.06		1.20	1.23	1.25	1.19	1.19	1.13	1.10
10	1.07	1.10	1.12	1.06	1.05	1.00	0.97		1.02	1.04	1.06	1.02	1.01	0.96	0.93		1.11	1.14	1.15	1.10	1.10	1.05	1.02		1.15	1.18	1.21	1.15	1.14	1.09	1.06
11	1.05	1.08	1.10	1.04	1.03	0.98	0.95		1.00	1.03	1.04	1.00	0.99	0.94	0.92		1.09	1.12	1.13	1.08	1.08	1.03	1.00		1.13	1.16	1.18	1.13	1.12	1.07	1.04
12	1.05	1.09	1.10	1.04	1.04	0.99	0.96		1.01	1.03	1.05	1.00	1.00	0.95	0.92		1.10	1.13	1.14	1.08	1.09	1.03	1.01		1.14	1.17	1.19	1.14	1.13	1.07	1.04
13	1.02	1.05	1.07	1.01	1.01	0.95	0.93		0.97	1.00	1.02	0.97	0.97	0.92	0.89		1.06	1.09	1.10	1.05	1.06	1.00	0.97		1.10	1.13	1.15	1.10	1.10	1.04	1.01
14	1.01	1.04	1.06	1.00	1.00	0.95	0.92		0.96	0.99	1.01	0.96	0.96	0.91	0.88		1.05	1.08	1.09	1.04	1.05	0.99	0.97		1.09	1.12	1.14	1.09	1.08	1.03	1.00
15	1.02	1.05	1.07	1.01	1.01	0.95	0.93		0.97	1.00	1.02	0.97	0.97	0.92	0.89		1.06	1.09	1.10	1.05	1.06	1.00	0.97		1.10	1.13	1.15	1.10	1.09	1.04	1.01
16	1.01	1.04	1.05	1.00	0.99	0.94	0.92		0.96	0.98	1.00	0.96	0.95	0.90	0.88		1.05	1.08	1.08	1.03	1.04	0.99	0.96		1.09	1.12	1.14	1.08	1.08	1.02	1.00
17	0.99	1.02	1.04	0.98	0.98	0.93	0.91		0.95	0.97	0.99	0.95	0.94	0.89	0.87		1.04	1.06	1.07	1.02	1.03	0.98	0.95		1.08	1.10	1.12	1.07	1.07	1.01	0.99
18	1.01	1.04	1.06	1.00	0.99	0.94	0.92		0.96	0.99	1.01	0.96	0.96	0.91	0.88		1.05	1.08	1.09	1.04	1.04	0.99	0.96		1.09	1.12	1.14	1.09	1.08	1.03	1.00
19	1.04	1.07	1.09	1.03	1.03	0.98	0.95		1.00	1.02	1.04	0.99	0.99	0.94	0.91		1.09	1.12	1.13	1.07	1.08	1.02	1.00		1.13	1.16	1.18	1.12	1.12	1.06	1.03
20	1.05	1.08	1.10	1.04	1.04	0.98	0.96		1.00	1.03	1.05	1.00	1.00	0.94	0.92		1.10	1.12	1.13	1.08	1.09	1.03	1.00		1.14	1.17	1.19	1.13	1.13	1.07	1.04
21	1.05	1.08	1.10	1.04	1.03	0.98	0.95		1.00	1.02	1.04	0.99	0.99	0.94	0.91		1.09	1.12	1.13	1.08	1.08	1.03	1.00		1.13	1.16	1.18	1.13	1.12	1.06	1.04
22	1.03	1.06	1.08	1.02	1.02	0.97	0.94		0.99	1.01	1.03	0.98	0.98	0.93	0.90		1.08	1.10	1.11	1.06	1.07	1.01	0.99		1.12	1.15	1.17	1.11	1.11	1.05	1.02
23	0.98	1.01	1.03	0.97	0.97	0.92	0.89		0.94	0.96	0.98	0.93	0.93	0.88	0.86		1.02	1.05	1.06	1.01	1.02	0.96	0.94		1.06	1.09	1.11	1.06	1.05	1.00	0.97

Appendix I: Descriptive Statistics of Monitored and Modelled Indoor Concentrations for Home A

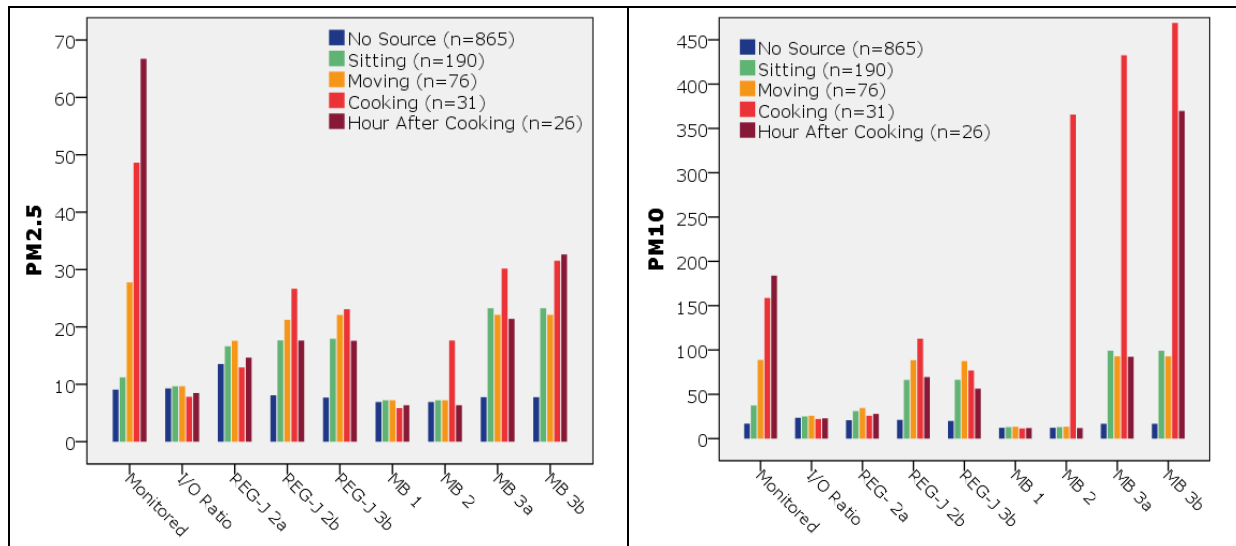
Appendix I, Table 1: Descriptive statistics of hourly indoor modelled concentrations at Home A in comparison to monitored indoor concentrations for PM_{2.5} and PM₁₀ (in µg/m³). Model types applied are: indoor-outdoor (I/O) ratio, regression models (REG-J) and mass-balance models (MB); for further description of the models, see chapter 5.

	Model	Mean	Median	Standard Deviation	Range
PM _{2.5}	Monitored:	12.89	8.12	38.56	838.16
	I/O Ratio	9.30	8.26	5.54	62.24
	REG-J 2a	14.30	11.51	10.27	81.28
	REG-J 2b	11.17	9.07	7.44	41.32
	REG-J 3b	10.89	8.36	7.39	49.28
	MB1	6.95	6.16	4.14	46.48
	MB2	7.26	6.35	4.77	46.48
	MB3a	12.06	8.38	11.48	81.12
	MB3b	12.34	8.39	11.98	81.12
PM ₁₀	Monitored:	32.04	14.64	93.89	2020.48
	I/O Ratio	23.83	21.84	13.91	222.95
	REG-J 2a	23.60	17.74	20.43	194.65
	REG-J 2b	36.10	24.31	33.84	254.57
	REG-J 3b	33.93	22.72	31.95	269.96
	MB1	12.36	11.33	7.21	115.60
	MB2	21.80	11.62	75.76	1062.27
	MB3a	47.46	15.22	99.12	1157.33
	MB3b	54.61	15.23	118.83	1157.33

Indoor Mean Concentrations by Source Related Situation

A comparison of the mean concentration for each source related indoor situation are presented in a bar chart in Appendix I, Figure 1Error! Reference source not found.. The selection of source categories for the graphs generally follows the differentiation undertaken by Jamieson’s regression model 2b (sitting, moving, and cooking). Sleeping was however included in the “no source activities” category for simplification. “Hour after cooking” has been added as a category. The number of hours monitored (and modelled) for each category is given as *n*.

Appendix I, Figure 1: Mean concentrations per source related situation for monitored and modelled PM_{2.5} and PM₁₀ at Home A. Models include: indoor-outdoor (I/O) ratio, regression (REG-J) models, and mass-balance (MB) models; for further description of the models, see chapter 5.



Appendix J: Descriptive Statistics of Ambient Monitoring Stations Used for Temporal Adjustments of the LUR Model as Part of the Personal Exposure Model

Appendix J, Table 1: PM_{2.5} concentrations (µg/m³) at background monitoring stations applied as part of the temporal adjustment of the land-use regression model for the integration into the MEPEX model.

PM _{2.5}	n	Mean	SD	Range
Bexley, Belvedere	539	9.26	6.20	58
Harrow, Stanmore	882	12.01	8.18	73
Hillington, Harlington	842	13.24	9.88	121
Kensington and Chelsea, North Kensington	1025	13.14	8.51	71
Camden, Bloomsbury	831	14.86	9.22	70
Average	1055	12.56	7.70	62.60

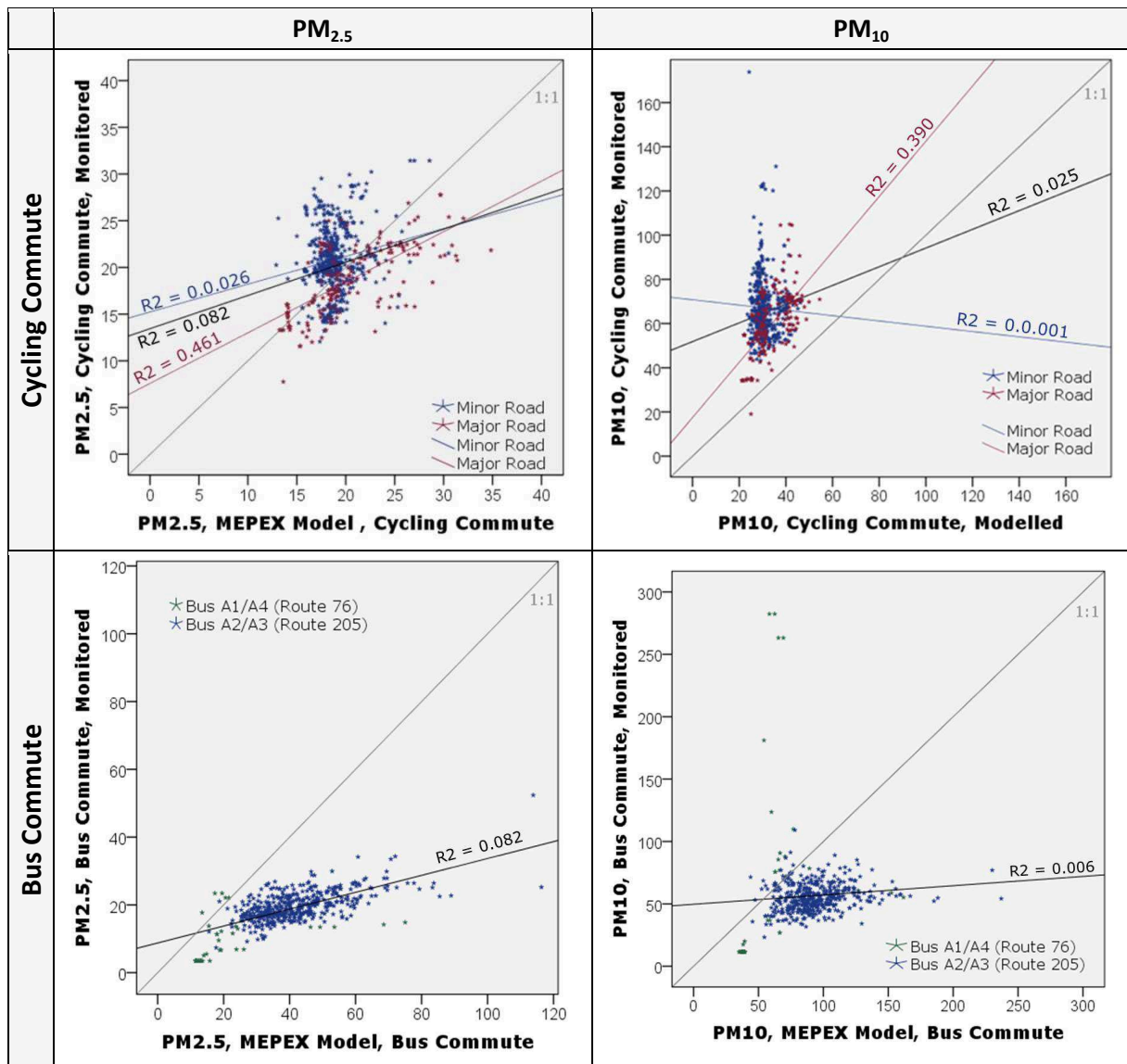
Appendix J, Table 2: PM₁₀ concentrations (µg/m³) at background monitoring stations applied as part of the temporal adjustment of the land-use regression model for the integration into the MEPEX model.

PM ₁₀	n	Mean	SD	Range
Camden, Bloomsbury	985	17.92	10.73	81
City of London, Sir John Cass School	972	28.66	16.57	226
Westminster, Horseferry Road	571	18.24	7.93	44
Haringey, Priory Park South	1053	18.1	12.04	219
Barnet, Finchley	1042	19.38	9.33	89
Tower Hamlet, Poplar	763	22.58	10.60	91
Newham, Wren Close	816	24.33	11.04	76
Islington, Arsenal	1035	21.42	11.36	143
Lambeth, Streatham Green	930	24.14	27.51	495
Kensington, Chelsea, North Kensington	993	20.42	10.41	85
Brent, St Mary's Primary School	663	20.25	9.35	66
Waltham Forest, Dawlish Road	825	22.77	10.26	88
Lambeth, Loughborough Junction	592	20.42	11.84	69
Average	1055	20.82	9.35	72.30

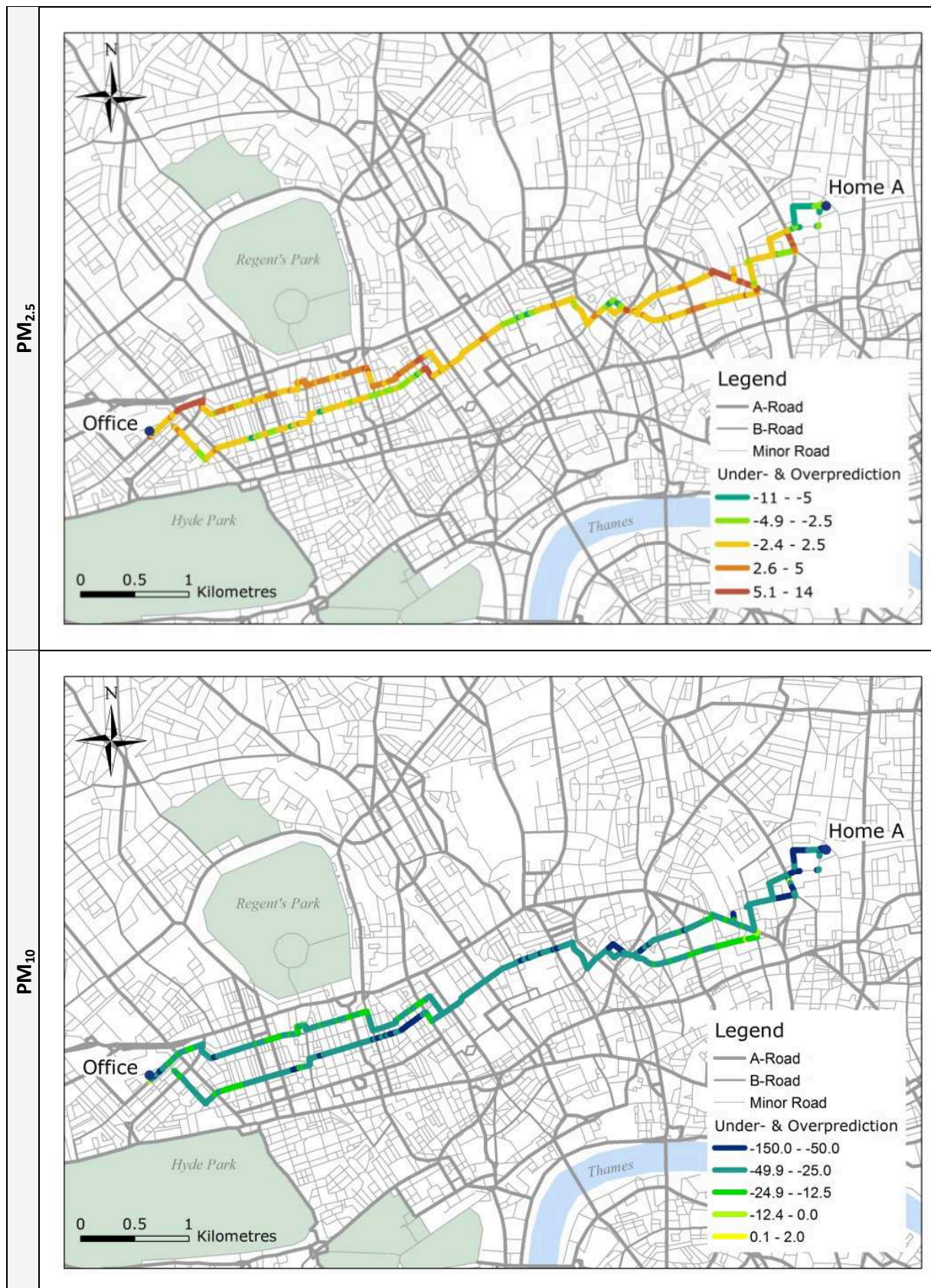
Appendix K: Modelled and Monitored Cycling and In-Bus Particle Exposure of Individual A along Segments of Commuter Routes

In this section a detailed investigation of the difference between monitored and modelled concentrations along the cycling and bus commutes routes of individual A is undertaken. The LUR-adj modelled concentrations, as well as monitored concentrations attached to GPS points have been aggregated for 20m road segments along the route (as described in chapter 5 for transport mode comparisons). For this analysis all road segments of morning and evening commute of each transport mode are evaluated together. Scatter plots for modelled against monitored concentrations are presented in Appendix K, Figure 1 **Error! Reference source not found.**. The scatter plot for cycling is differentiated by road segments along minor and major roads, according to the categories used by the LAEI ('A roads' and 'B roads' are considered major roads, 'Minor roads' are considered minor roads). The scatter plots for the bus commute are differentiated by two different bus routes taken along the commute. Maps with modelled minus monitored concentrations along the commute are presented for $PM_{2.5}$ and **Error! Reference source not found.** for PM_{10} (see Appendix K, Figure 2 and Appendix K, Figure 3).

Appendix K, Figure 1: Modelled against Monitored PM_{2.5} and PM₁₀ concentrations (both in µg/m³) for the cycling and bus commutes of individual A, averaged by 20m road segment.



Appendix K, Figure 2: Cycling commute of individual A with monitored minus modelled PM_{2.5} and PM₁₀ concentrations (in $\mu\text{g}/\text{m}^3$) aggregated for 20m long segments along the route.



Appendix K, Figure 3: Bus commute of individual A with monitored minus modelled PM2.5 and PM10 concentrations (in $\mu\text{g}/\text{m}^3$) aggregated for 20m long segments along the route.

