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Assessing the contributions of outdoor and indoor sources to air quality in London homes of the SCAMP cohort

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ABSTRACT

Given that many people typically spend the majority of their time at home, accurate measurement and modelling of the home environment is critical in estimating their exposure to air pollution. This study investigates the fate and impact on human exposure of outdoor and indoor pollutants in London homes, using a combination of sensor measurements, outdoor air pollution estimated from the CMAQ-urban model and indoor mass balance models. Averaged indoor concentrations of $PM_{2.5}$, PM_{10} and NO_2 were 14.6, 24.7 and 14.2 μg m^{-3} while the outdoor concentrations were 14.4, 22.6 and 21.4 μg m^{-3}, respectively. Mean infiltration factors of particles (0.6–0.7) were higher than those of NO_2 (0.4). In contrast, higher loss rates were found for NO_2 (0.5–0.8 h^{-1}) compared to those for particles (0.1–0.3 h^{-1}). The average concentrations of $PM_{2.5}$, PM_{10} and NO_2 in kitchen environments were 22.0, 33.7 and 20.8 μg m^{-3}, with highest hourly concentrations (437, 644 and 136 μg m^{-3}, respectively) during cooking times (6–7 pm). Indoor sources increased the indoor concentrations of particles and NO_2 by an average of 26–37% in comparison to the indoor background level without indoor sources. Outdoor and indoor air exchange plays an important role in reducing air pollution indoors by 65–86% for particles and 42–65% for NO_2.

1. Introduction

People spend a major part of their time indoors, and this is especially true for children and vulnerable groups [1–3]. According to the US National Human Activity Pattern Survey, people spend approximately 68% of their time at home indoors [4]. A report by the Royal College of Paediatrics and Child Health found that children in the UK only play around 1 h per day outdoors, and 3.6 million children (35% of the number of UK pupils) are living in poor quality housing [5]. Consequently, they may experience greater exposure to air pollution indoors than outdoors, and the home environment and its pollutant sources play important roles in determining human exposure to air pollution [6].

According to the Global Burden of Disease Study, exposure to household air pollution due to the use of solid fuels accounted for 1.8 million (95% CI 1.1–2.7) deaths and 60.9 million (34.6–93.3) disability-adjusted life years (DALYs) while ambient ozone and $PM_{2.5}$ were attributed to 3.1 million deaths and 86.1 million DALYs in 2017 globally [7,8]. Lee at al [7]. conducted a meta-analysis of health impacts of household air pollution and found that household air pollution was high positively associated with respiratory and cardiovascular illnesses such as asthma (RR 1.23, 95% CI 1.11–1.36), acute respiratory infection in both adults (1.53, 1.22–1.93) and children (1.39, 1.29–1.49), chronic obstructive pulmonary disease (1.70, 1.47–1.97), lung cancer (1.69, 1.44–1.98), and cardiovascular (1.07, 1.04–1.11) mortality. Therefore, apportionment of the contribution of both outdoor and indoor sources to home air quality is critical in estimating human exposure to air pollution and associated health outcomes. It is also an essential step for risk assessment and control of indoor air quality.

Home air quality is not only controlled by its indoor sources, but also by the ingress of outdoor air pollutants [9,10]. Indoor air pollutants can be generated from human activities such as smoking, cleaning, ironing and cooking [11–13], or can be formed as secondary pollutants from

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indoor chemistry, such as secondary aerosols from indoor gas-to-particle conversion [14]. When a pollutant enters indoors from outdoors, it is subjected to indoor dynamic processes such as dilution, dispersion, coagulation, evaporation, deposition and chemical reactions including the reactions between ozone, VOCs and NO [13,15]. The ingress of outdoor air pollution, sinks and sources are represented by infiltration factors, loss rates and indoor emissions rates - which are key parameters in indoor modelling. These parameters are also strongly influenced by the air exchange rate, the characteristics of each pollutant species and building characteristics. Koivisto et al. [6] emphasized that good information on emission sources in combination with well parameterized mass balance models could give us comprehensive picture of factors influencing indoor environments.

London is home to more than 9 million people who live in areas where the level of fine particles exceeded the new 2021 WHO guideline of 5 μg m^{-3} [16]. Although outdoor air pollution in London has been extensively investigated through numerous experimental and modelling studies in recent decades [17], very few have been designed to focus on indoor air quality in home environments, especially for homes with child occupants. Previous measurement and modelling studies on indoor air quality in London homes includes Melia et al. [18] who measured weekly average NO_2 in both outdoor and indoor environments in 29 London homes, and Smith et al. [19] who developed a London Hybrid Exposure Model (LHEM) to estimate human exposure at home indoors (but without indoor sources). However, one of the limitations of those studies is that key parameters for indoor modelling such as ventilation, infiltration factors and loss rates of air pollutants were not comprehensively investigated. Furthermore, the contribution of indoor and outdoor sources to indoor air quality at London homes were not apportioned. More data on indoor levels of particles and gaseous pollutants, and air exchange rates in London homes, are therefore required.

The objectives of this study were: i) to determine the infiltration factors of $PM_{2.5}$, PM_{10} and NO_2; ii) to quantify the role of ventilation and indoor dynamics in reducing indoor air pollution; and iii) to calculate the diurnal pattern of indoor sources contributions to indoor air quality at homes. A mass balance model [20] was applied to 132 London homes, using data sets of outdoor air quality, modelled by the CMAQ-urban model [21], and indoor air quality measured using pollution sensors.

2. Methodology

2.1. Experimental design

2.1.1. Data collection

This work is nested within the SCAMP study – a cohort study of secondary school children in Greater London [22]. Indoor and outdoor air pollution data were collected for 132 London homes (of SCAMP study participants-with child occupants aged 11–15 years) over 3 years from 2016 to 2018. The pollution monitors were placed at each house for a minimum of 48 h as part of the MRC-funded Cognitive Development in the Urban Environment (CLUE) project. Hourly concentrations of particulate matter ($PM_{2.5}$ and PM_{10}) and gaseous pollutants (NO_2 and CO) were measured simultaneously in both outdoor and indoor environments of each home. Unfortunately, each home had only one observation in either the warm or cold season due to limited resources (i.e. a small number of sensors). This could make it difficult to investigate the impact of background pollution of ambient environments on the indoor data between homes and to compare seasonality of both outdoor and indoor air quality. Future studies ought to be designed with longer and synchronized sampling campaigns for each home (i.e. in both winter and summer seasons) to overcome the limitations of our study.

PM measurements were made using two types of optical sensors (PurpleAir and TZOA) which detect particles based on light-scattering technology. The TZOA sensor (research model- RD02, manufactured by TZOA WearablesTM, Canada) has three non-overlapping size bins with cut-point of 1, 2.5 and 10 μm and a particle size detection limit of 0.3 μm [23]. The PurpleAir (PA-II unit, PurpleAir Ltd, US) contains two PMS5003 sensors which count the pulses from scattering signal and then convert number of pulses to a mass concentration of particles at six cut-points with a diameter range of 0.3–10 μm [24]. The PurpleAir sensor can measure PM in the range from 0 to 500 μg m^{-3} with maximum consistency error of 10 μg m^{-3}. The total response time is 10 s. A note is that particles mainly dominate in the size range lower than 0.3 μm in number but have very low contribution to PM mass in London [25]. The gaseous pollutants were measured using AQMesh devices (Environmental Instruments Ltd, UK, www.aqmesh.com) which use electrochemical sensors to measure pollutants. Measurement range of AQMesh devices was 0–20000 ppb with Limit of detection lower than 1 ppb for NO_2 and O_3. Time resolution for measurements was every minute. We aggregated all measured data into hourly time resolution in the final version for data analysis. In terms of indoor measurements, the devices were deployed in the main communal area of the house - either the living rooms (104 measurements) or the kitchens (28 measurements). The sensors were placed in breathing zones within communal areas, and avoided blocked locations. As with many low-cost sensors, the quality of readings diminished over time, and the sensors were replaced when required, as identified via the QA/QC regime. A total of 9 AQMeshs, 5 TZOAs and 6 PurpleAirs devices were used during the sampling campaign, be deployed sequentially at the 132 homes.

2.1.2. Quality assurance and quality control

The sensors were co-located at an urban roadside (Marylebone Road) site (https://uk-air.defra.gov.uk/networks/site-info?site_id=MY1) for separate week-long periods before, during and after the measurement campaign. At London Marylebone monitoring stations, hourly concentrations of PM were measured by the Thermo TEOM 1400AB/8500 FDMS (Filter Dynamic Measurement System while hourly concentrations of NO2 were measured by chemilumninescence methods.

Deming regression was used to calibrate the sensors' readings after co-location with reference instruments. The advantage of using this linear regression technique over the ordinary least squares method is that it takes into account measurement error in both the dependent and independent variables [26].

A slope, intercept and R^2 values were calculated for each co-location, pollutant and individual sensor. Where the R^2 was less than 0.5 these co-location correction factors were discarded. This amounted to 31 out of 171 total co-locations being removed. Additionally, the co-locations for NO_2 were removed as the R^2 were all less than 0.65 and slopes and intercepts were substantially different compared to other co-location periods; this resulted in a further 24 co-locations being discarded. The remaining co-locations were averaged for the slope and intercept of each individual sensor and were used to correct raw measured values. After the QA/QC processes were completed, measurements in 72, 71 and 81 homes for $PM_{2.5}$, PM_{10}, NO_2 remained respectively. A summary of correction factors is shown in Table S1.

2.2. Outdoor and indoor modelling framework

2.2.1. Outdoor modelling using CMAQ-Urban model

To predict detailed outdoor air pollution, we coupled the Weather Research and Forecasting (WRF) meteorological model v3.6.1 (National Centre for Atmospheric Research, Boulder, CO), and Community Multiscale Air Quality (CMAQ) model v5.0.2 (U S. Environmental Protection Agency, Washington, DC), with the Atmospheric Dispersion Modeling System (ADMS) Roads model v4 (Cambridge Environmental Research Consultants, Cambridge, UK) [21,27]. We refer to this model as CMAQ-urban.

CMAQ-urban used anthropogenic emissions data, obtained by combining the UK National Atmospheric Emissions Inventory (NAEI), the London Atmospheric Emissions Inventory, Imperial College's road transport emissions model, and the European Monitoring and Evaluation Programme (EMEP) emissions. The biogenic emissions from

vegetation and soils were estimated using the Biogenic Emission Inventory System version 3 (BEIS3) model (U.S. Environmental Protection Agency) and sea-salt emissions were calculated in line with CMAQ [21]. The CMAQ-urban model outputs hourly air pollution concentrations at 20 m grid resolution across the study domain. The model predicts $PM_{2.5}$, PM_{10}, nitrogen oxides (NO_X), NO_2 and O_3, with the ADMS-Roads model used to describe near field dispersion from roadways, and a simple NO, NO_2 and O_3 chemical scheme employed [28,29].

In this study, hourly outdoor concentrations of $PM_{2.5}$, PM_{10} and NO_2 at London homes were extracted from the CMAQ-Urban model every 20 m. This data was used to compare the corresponding outdoor concentrations of pollutants measured by the deployed sensors placed outside the home. Unexpectedly high outdoor concentrations of PM and NO_2 were measured by the sensors at 21 homes during cooking times (when high peaks of particles or NO_2 occurred during 5–8 pm). It was concluded that the data were strongly affected by indoor sources owing to the sensors' proximity to kitchen windows, and measured data were therefore replaced with that from the CMAQ-Urban model. Outdoor measurement data was also classified as being of poor quality if it was: 1) lower than indoor concentrations during sampling campaign or 2) having peaks at cooking times that are lower than those peaks from concurrent indoor measurements. Correlations (r) between sensor measurements (after removing suspected and poor data) and the CMAQ-Urban model were 0.84, 0.69 and 0.64 for $PM_{2.5}$, PM_{10} and NO_2 as shown in Fig. S1.

2.2.2. Indoor modelling framework

The indoor modelling framework is shown in Fig. 1. First, our algorithm identifies the indoor source events based on the indoor concentrations of both particles ($PM_{2.5}$, PM_{10}) and gaseous pollutants (NO_2, CO). Second, air exchange rates are estimated using the decay rate of CO after indoor source events, as well as infiltration factors and loss rates using a dynamic mass balance model. Third, these estimated parameters are used to predict indoor concentrations of air pollutants without indoor sources. Finally, modelled and measured indoor concentrations of pollutants are compared in the absence of indoor sources, allowing the contribution of indoor sources to indoor concentrations of pollutants to be apportioned [30].

2.2.3. Air exchange rate estimation

In our study, we estimate air exchange rate (AER) as a decay rate of indoor carbon monoxide (CO) after an indoor source of CO occurs.

The following criteria were used to identify peaks in indoor concentrations due to indoor sources: 1) The peak concentration was at least three times higher than the averaged concentration of indoor concentrations over the sampling period; and 2) The peak concentration was higher than outdoor concentrations at the same time and previous time (hour) point; and 3) the indoor concentration at the time point was twice as high as during the previous hour, and showed a decrease in concentration over the next 6 h. A time period of 6 h after the peak concentration was used to estimate the AER values (α) using equation (1):

$$CO_{in\ (at\ ime\ point\ t)} = CO_{\text{in}\ back-ground} + CO_{\text{in}-(at\ peak\ time+1h)}\ e^{(\alpha+k_{d-CO})*t} \quad (1)$$

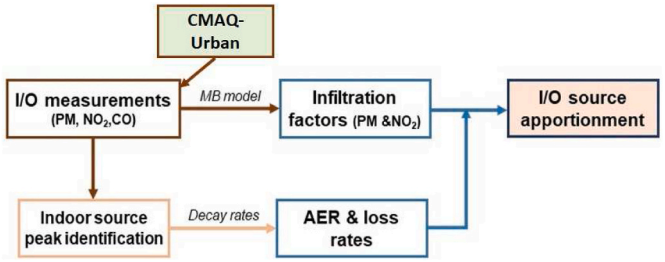


Fig. 1. Indoor modelling framework.

In equation (1), α is the air exchange rate (h^{-1}) and $k_{d\text{-}CO}$ is deposition rate of CO. Because carbon monoxide can be considered as an unreactive gas, the deposition rate of CO ($k_{d\text{-}CO}$) is assumed to be zero [31,32]. The fitting was over the period from the hour after peak concentration to 6 h after the event, using a self-starting asymptotic regression from the Nonlinear Least Squares (nls) R package. It was assumed that the air had been well-mixed indoors between living room/kitchen 1 h after the end of the indoor pollution event. R^2 obtained from above fittings was ranged from 0.63 to 0.96.

2.3. A dynamic mass balance model to model indoor air quality from outdoors

Assuming that the indoor air is well mixed, the indoor concentration of a pollutant can be estimated using a numerical backward difference for each time step of the specified period (Δt) [33]:

$$C_{in\ (t+\Delta t)} = \frac{\alpha * P * C_{out\ (t+\Delta t)}}{\alpha + k}\left(1 - e^{-(\alpha+k)*\Delta t}\right) + C_{in\ (t)}e^{-(\alpha+k)*\Delta t} + \frac{S_{t+\Delta t}}{(\alpha+k)*V}\left(1 - e^{-(\alpha+k)*\Delta t}\right) \quad (2)$$

in which P and k are the penetration factor and loss rate (due to deposition, evaporation or chemical reactions) of a pollutant (h^{-1}), respectively. S represents the strength of each indoor source for each pollutant ($\mu g.h^{-1}$). V is the volume of the room (m^3). The time step between two indoor and outdoor measurements (Δt) is 1 h.

The infiltration factor (F_{in}) is defined as the equilibrium fraction of outdoor particles that penetrates indoors and remains suspended; it could therefore be estimated as below [33]:

$$F_{in} = \frac{\alpha * P}{\alpha + k} \quad (3)$$

The penetration factor is unity for gaseous pollutants, while it is lower than 1 for particulate matter. Hence, F_{in} is lower than 1 for both particles and NO_2. Combined with the equation (3), the equation (2) can be transformed to the following equation:

$$C_{in\ (t+\Delta t)} = F_{in}C_{out\ (t+\Delta t)}\left(1 - e^{-(\alpha+k)*\Delta t}\right) + C_{in\ (t)}e^{-(\alpha+k)*\Delta t} + C_{in\ by\ IS\ (t+\Delta t)} \quad (4)$$

Indoor source events of NO_2 and particles were identified in an analogous way to those of carbon monoxide. The indoor peaks were discarded from the dataset, and a mass balance model based on equation (4) was used to estimate the infiltration factors and loss rates of pollutants for each time period of 6 h after an indoor source event ($S = 0$).

2.4. Infiltration factor and loss rate estimation

To determine the infiltration factor and loss rate (F_{in}, k), the F_{in} and k values (with $0 < F_{in} \leq 1$ and $k > 0$) were varied until the modelled C_{in}, using a mass balance model (equation (4)), had the best fit with the C_{in} measured by sensors. Goodness-of-fit was selected based on the root mean square error (RMSE) and the Pearson correlation coefficient (r) between the indoor data sets estimated by the model and observed by sensor measurements [20].

A dynamic mass balance model was applied to both the night-time and daytime periods without a peak concentration from indoor sources. In the case of no indoor source, equation (4) can be transformed to equation (5) below:

$$C_{in\ (t+\Delta t)} = F_{in}C_{out\ (t+\Delta t)} + \left(F_{in}C_{out\ (t+\Delta t)} - C_{in\ (t)}\right)e^{-(\alpha+k)*\Delta t} \quad (5)$$

If $C_{in\ (t)}$ is equal to $F_{in}C_{out\ (t+\Delta t)}$ (steady state conditions), indoor concentrations modelled by equation (5) strongly depend on infiltration factor rather than loss rates.

2.5. Estimation of the increment of indoor concentrations by indoor sources

Based on the values of infiltration factors and loss rates of air pollutants, a dynamic mass balance model was used to model indoor concentration in the case of no indoor sources following equation (5) over the whole the sampling period (we refer to the modelled indoor concentration as $C_{\text{in-model without indoor sources}}$). For the periods during which sources occurred, we represented the infiltration factors and loss rates by calculating the respective averages over the 6 h before and after each peak. An increment of indoor concentrations of air pollution associated with indoor sources was estimated by subtracting the concentrations predicted by our mass balance model from the indoor concentrations measured by sensors:

$$C_{In\ by\ IS\ (t)} = C_{In\ by\ sensor\ measurement\ (t)} - C_{In\ by\ model\ without\ indoor\ source\ (t)} \quad (6)$$

The limitation of this estimation is that infiltration factors and total loss rates (air exchange rate and deposition/evaporation rates) are dynamic, therefore our assumption of fixed numbers of infiltration factors could cause an uncertainty in our estimation. However, it is not easy to directly measure infiltration factors and air exchange rates during indoor source events. Such measurements are out with this study scope and could be in the future studies.

3. Results

3.1. Overview of indoor/outdoor air quality at London homes

As shown in Table 1, averaged concentrations of $PM_{2.5}$, PM_{10} and NO_2 were 14.6, 24.7 and 14.2 μg m^{-3} indoors, while those outdoors were 14.4, 22.6 and 21.4 μg m^{-3}, respectively. The levels of these air pollutants were within the EU annual mean ambient air quality limits of 25, 40, and 40 μg m^{-3} for $PM_{2.5}$, PM_{10} and NO_2 respectively. However, outdoor concentrations of PM and NO_2 exceeded the new 2021 WHO annual mean ambient air quality guideline by 1.5–3 times (5, 15 and 10 μg m^{-3} for $PM_{2.5}$, PM_{10} and NO_2 respectively).

Mean Indoor/Outdoor (I/O) ratios were 1.22 for $PM_{2.5}$ and 1.21 for PM_{10}, whereas for NO_2 the ratio was 0.84. Fig. 2 shows diurnal patterns of hourly I/O ratios of PM and NO_2. A distinct peak in PM is observed at 6–7 pm and two smaller peaks are observed in the morning and mid-afternoon, corresponding to cooking times for London's households. There is a distinct peak for I/O ratios of NO_2 at 10 a.m., and higher ratios were found during the daytime.

Highest I/O values of NO_2 not only depend upon the indoor sources, but also outdoor sources. At 10 a.m., outdoor concentration of NO_2 dropped quickly due to huge reduction of traffic load (traffic rush hour in London is around 7–9 am), but the indoor concentration of NO_2 decreased more slowly. It made the highest I/O value of NO_2 at 10 a.m. At 4 a.m., the peak is due to lower outdoor concentration NO_2 at this time as shown in Fig. 6, whereas there was an increase of indoor NO_2 from 4 to 7 p.m. due to cooking emissions. The difference between I/O ratios of PM and NO_2 is not only due to difference between indoor sources of PM and NO_2 but also due to time-series changes in PM and NO_2 concentrations. For example, cooking by gas emit both PM and NO_2 while electric cooking or resuspension mainly release indoor PM, not NO_2. A quicker drop of outdoor NO_2 concentrations after traffic rush hours is probably due to chemistry of outdoor NO_2 in the reaction system of NO–NO_2–O_3 during day-time.

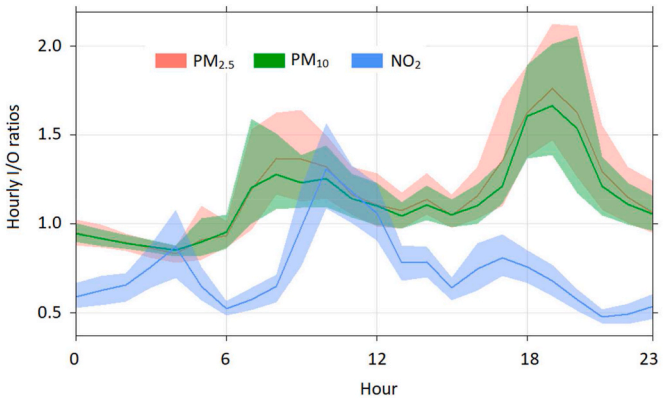


Fig. 2. Diurnal patterns of hourly I/O ratios of $PM_{2.5}$, PM_{10} and NO_2. The line shows the mean concentrations and shaded area as 95% confidence interval in the difference in mean concentrations.

3.2. CO levels and air exchange rates

The average concentration of CO indoors was 0.45 ppm, with a typical peak range of 0.5–2 ppm at 7 p.m. which corresponds to the evening cooking times. Indoor levels of CO were much higher than those outdoors (0.03 ppm); median I/O ratios were typically larger than 4.0. This indicated that decay curves of CO after events of indoor sources could be utilized to estimate AER values using equation (1). As a result, Fig. 3 shows the distribution of estimated values of AER during summer and winter. The estimated AER values ranged from 0.1 to 2.9 h^{-1} with an average of 0.6 h^{-1}. These estimated values of air exchange rates are in agreement with previous studies. According to a database of 471 dwellings in UK, the mean AER is 0.7, with a range of 0.2–1.5 [34,35].

3.3. Infiltration factors and loss rates of indoor air pollutants

Fig. 4 shows density distributions of infiltration factors of PM and NO_2 during warm and cold seasons. Mean values of F_{in} for $PM_{2.5}$, PM_{10} and NO_2 were 0.74, 0.75 and 0.44 respectively in the warm season (May–October), which were higher than those in the colder season (0.53, 0.62 and 0.40 for $PM_{2.5}$, PM_{10} and NO_2, respectively). In addition, distributions of infiltration factors of PM show a peak of 0.82–0.84 in

Table 1
Statistics of indoor, outdoor concentrations and I/O ratios of PM and NO_2.

Pollutants	No of homes	Environment	Min.	1st Q	Median	Mean	3rd Q	Max
$PM_{2.5}$ (μg m^{-3})	72	In	1.2	7.3	9.6	14.6	14.1	437.2
		Out	1.5	7.2	9.7	14.4	15.4	272.7
		Ratio	0.07	0.70	0.93	1.22	1.12	33.4
PM_{10} (μg m^{-3})	71	In	1.9	13.9	17.2	24.7	22.6	644.4
		Out	1.5	14.7	17.3	22.6	24.4	219.5
		Ratio	0.1	0.81	0.95	1.21	1.10	35.86
NO_2 (μg.m^{-3})	81	In	0.2	7.8	11.6	14.2	16.8	136.0
		Out	0.2	12.6	20.0	21.4	29.2	92.6
		Ratio	0.01	0.39	0.58	0.84	0.89	9.6
CO (μg.m^{-3})	89	In	0.01	0.17	0.34	0.54	0.59	16.2
		Out	0.01	0.04	0.07	0.11	0.14	1.01
		Ratio	0.06	1.84	4.93	10.7	9.83	772

Min., 1st Q, 3rd Q and Max stand for minimum, 1st quarter, 3rd quarter and maximum respectively.

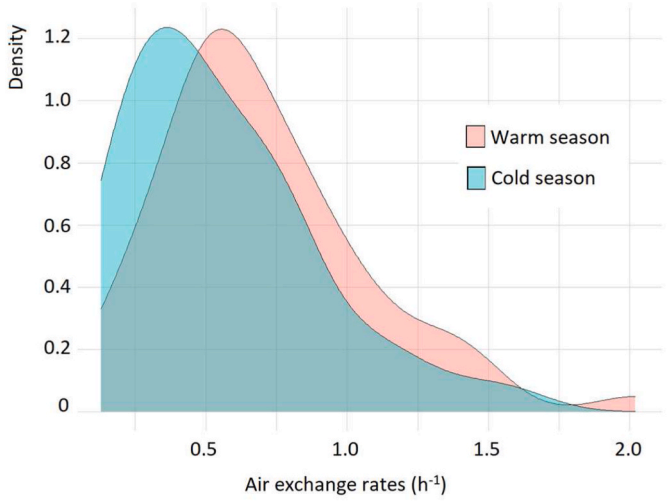


Fig. 3. Density distribution of air exchange rates between cold and warm seasons.

warm and 0.36–0.62 in cold season. The density distribution of infiltration factors NO_2 shows a unique peak of 0.33 in winter whereas it shows a bi-modal with two peaks at 0.25 and 0.5 in summer. Higher values in summertime could be due to increased air exchange rate in summer (0.70) compared to winter (0.53).

The mean loss rates (by deposition/evaporation/chemistry and air exchange) were 0.95, 0.82 and 1.25 h^{-1} for $PM_{2.5}$, PM_{10} and NO_2, respectively as shown in Fig. 5. After removing the loss by air exchange, loss rates of these pollutants due to their physico-chemical dynamics were 0.3, 0.25 and 0.75 h^{-1} for $PM_{2.5}$, PM_{10} and NO_2 respectively in winter; these values were slightly higher than those in summer (0.24, 011, and 0.5 h^{-1} for $PM_{2.5}$, PM_{10} and NO_2, respectively). There were not much different between density distribution of total loss rate of indoor air pollutants between cold and warm seasons. It is due to complex processes that influence on the loss or production of indoor pollutants between seasons. For example, the air exchange rate was lower in winter, but loss of PM could be higher in heating season due to evaporation of NH_4NO_3 indoors.

3.4. Contribution of indoor sources to indoor air quality

Having applied equation (6), Fig. 6 illustrates the temporal contribution of indoor sources to indoor air quality. Mean and median contributions of indoor sources to indoor concentrations of NO_2 were 37 and 33% respectively. In terms of particles, averages of 31% and 28% of indoor $PM_{2.5}$ and PM_{10} respectively were generated from indoor sources across the whole sampling period. The indoor contribution not only varied among homes but also by time of day. During the typical cooking time (at 6–7 pm), indoor sources (which were predominantly gas cookers/stoves) could increase indoor concentration of NO_2 by approximate 9.6 μg m^{-3}, accounting for of 67% of indoor NO_2. At 6–7 pm, indoor sources could increase the $PM_{2.5}$ and PM_{10} concentrations by an average of 16 and 21 μg m^{-3} respectively.

4. Discussion

4.1. Effects of seasonality and building characteristics on ventilation and indoor dynamics

The median value of AER during summer was 0.70 h^{-1}; this was slightly higher than that during winter (0.53 h^{-1}). The difference can be explained by the difference in outdoor/indoor climate and in human behaviour (i.e opened/closed windows) between the two seasons. A wider and bi-modal distribution during a warm season suggests that a higher occurrence of window opening leads to an increase in natural air exchange rate. Hill et al. [36] measured AER based on the decay of sulfur hexafluoride (SF_6) from 7 homes in northern England and reported that the mean AER for 13 periods with windows open was 1.37 air changes per hour, whereas for 17 periods with windows closed this value was nearly halved (0.79). A study by Crump et al. [37] conducted a two-week measurement campaign of air exchanges using a perfluorocarbon tracer in 37 new homes in England (built since 1995) and reported that a higher ventilation rate was found in summer (0.57) than in winter (0.43).

Air exchange rates also strongly depend upon the home's characteristics. As shown in Fig. S2, median AER values were highest for apartments (0.72 h^{-1}), followed by terraced houses (0.54 h^{-1}), semi-detached houses (0.51 h^{-1}) and detached houses (0.4 h^{-1}). However, large variation in AER is also observed within each type of building.

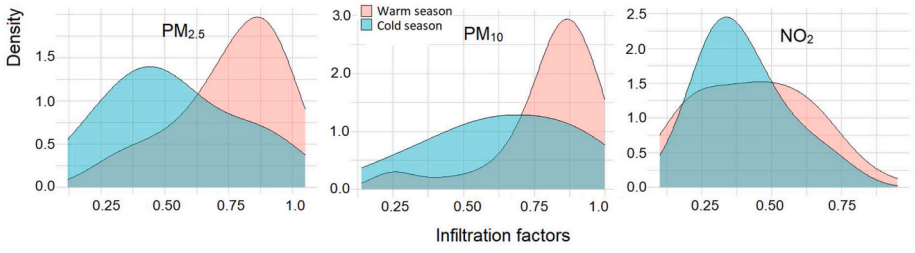


Fig. 4. Density distribution of infiltration factors between cold and warm seasons.

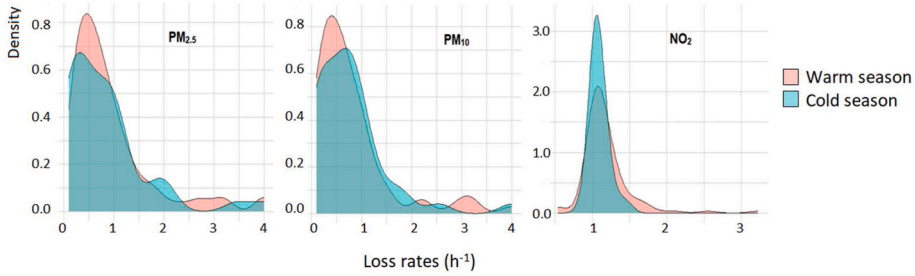


Fig. 5. Density distribution of total loss + AER rates of indoor air pollutants between cold and warm seasons.

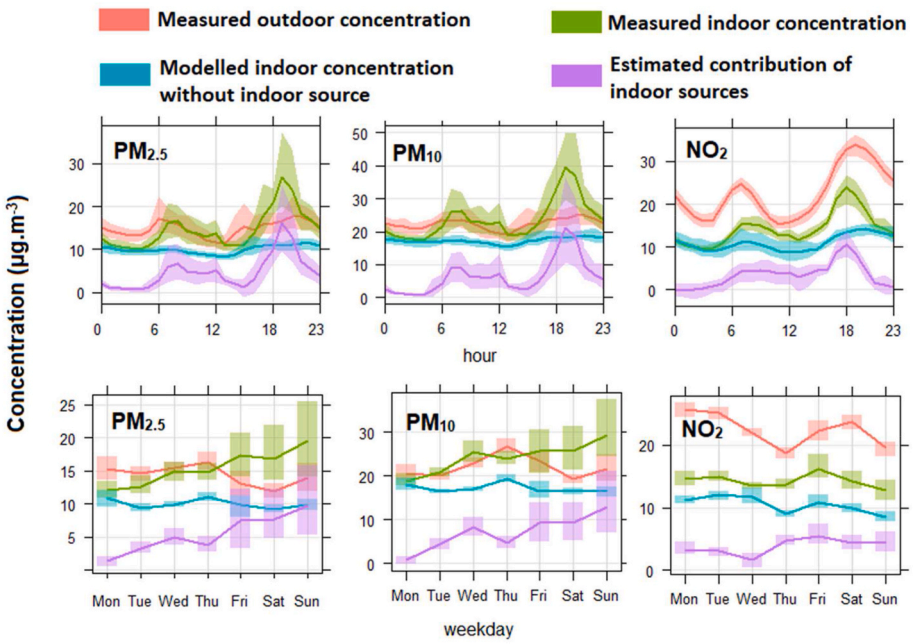


Fig. 6. Time variation of PM and NO_2, and indoor increments by indoor sources. The line shows the mean concentrations and shaded area as 95% confidence interval in the difference in mean concentrations.

Aizlewood and Dimitroulopoulou [38] reported air exchange rates measured from 8 flats in 2 apartment buildings ranging from 0.13 to 0.98, with values strongly depending upon whether trickle ventilators were open or closed.

Regarding indoor air pollution dynamics, infiltration factors for $PM_{2.5}$ were in the same range (0.6–0.7) as in many previous studies such as Meng et al. [39]; Ozkaynak et al. [40]; and Allen et al. [41]. Readers can refer to infiltration factors from a review by Chen and Zhao [33]. However, much lower values of F_{in} from 30 homes in Birmingham (UK) (mean values of F_{in} were 0.34 and 0.26 for $PM_{2.5}$ and PM_{10} respectively) were reported by Hoek et al. [42]. Different house characteristics among the studies is one likely reason for this, since some houses measured by Hoek et al. [42] used a mechanical ventilation system with air filters [33]. Additionally, as discussed above, infiltration factors of particles not only strongly depend upon their size but also their chemical composition. In another study, Jones et al. [43] used I/O ratios of sulphate (which can be used to represent for F_{in}) and found those values were 0.8, 0.7 and 0.6 for $PM_{1.0}$, $PM_{2.5}$ and PM_{10} respectively for seven homes in Birmingham (UK).

Like air exchange rates, the infiltration factors were found to be higher in summer. Based on equation (3), infiltration factor has an inverse relationship with ratios of deposition rate and air exchange rate (k/a), therefore an increase of air exchange rates may lead to an increase of infiltration factor. Other possible influencing factors include the change in chemical composition of aerosols between indoor and outdoor locations and transformations of aerosols between seasons, such as an increase of NH_4NO_3 loss due to I/O temperature gradient in winter [44]. In terms of loss rates of indoor air pollution, average air exchange rates were found to be higher than loss rates of particles due to deposition and evaporation. These results indicate that ventilation rate plays an important role in loss of indoor air pollutants, reducing indoor particles by 65–86% and NO_2 by 42–65%.

4.2. I/O ratios and contribution of indoor sources

High I/O ratios suggest a larger contribution of indoor sources to indoor concentrations of PM than NO_2 in London homes. Indoor sources of PM include combustion sources such as cooking and heating, but also non-combustion sources such as cleaning and resuspension from walking, while indoors NO_2 is generated mainly from gas cooking or heating in British homes [45]. Melia et al. [18] conducted measurements of NO_2 in 29 London homes of school children in winter, and found that homes with gas cookers and kerosene heaters had nearly double the NO_2 concentrations in their kitchens and living rooms when compared to homes without these sources. I/O ratios of NO_2 calculated here are consistent with a previous report by Dimitroulopoulou and Squire [46]; who found I/O ratios were in the range of 0.8–1.0 for kitchens and lounges from 35 new homes in England.

Comparison of I/O ratios with previous studies includes, an early work by Jones et al. [43]; who measured both indoor and outdoor PM in seven domestic homes within Birmingham (UK), reporting that mean I/O ratios ranged from 1.0 to 3.9, and were strongly dependent upon both indoor sources (cooking, smoking and resuspension) and ventilation rates. This is in agreement with a later study by Nasir and Colbeck [47]; who found mean I/O ratios were 2.1 and 1.5 for $PM_{2.5}$ and PM_{10} respectively. In contrast, a recent study by Stamp et al. [48] found lower I/O ratios (0.32–0.72 for $PM_{2.5}$ and 0.5–1.42 for PM_{10}) from a case study of five low-energy London apartments. These lower PM I/O ratios compared to this study and other previous studies may be explained by higher air exchange rates (0.7–1.5) in apartments and other factors, such as difference of strength in indoor sources among homes and where measurements were conducted.

This study found a significant contribution of indoor sources to indoor concentrations of both particles and NO_2, especially during typical cooking times (6–7 pm). Similar findings were reported by Dimitroulopoulou et al. [34] who developed a probabilistic model to predict air pollutant concentrations in UK homes. In particular, Dimitroulopoulou et al. .(2006) found that gas cooking can increase the concentrations of NO_2 in the bedroom, lounge, and kitchen by 2.6, 11.7, and 30.1 μg m^{-3}, 2.7, 8.4, and 19.2 μg m^{-3} for $PM_{2.5}$ and 5.5, 18.1 and 44.2 μg m^{-3} for PM_{10}, respectively. Jung et al. [49] who investigated both indoor and outdoor air quality obtained from 5000 homes (participants with child occupants at ages 1.5 and 3 years) in Japan reported I/O ratios of NO_2 were higher than 1, indicating significant contributions of indoor sources. It also found an increase of NO_2 at homes associated with operation times of gas heaters and gas ovens. In a recent review of I/O ratios of NO_2, Hu and Zhao [50] indicated that NO_2 emission rates would be very high in naturally ventilated residential

buildings in rural areas due to using traditional stoves for cooking.

In a comparison with other studies, Ji and Zhao [51] estimated indoor contribution was 4–45% of $PM_{2.5}$ in Beijing's home. Based on a recursive mass balance model combined with censoring algorithmes, Allen et al. [41] found indoor-generated concentrations of PM were 1.8 $\pm$ 1.8 μg m^{-3}, accounting for 22% of total indoor PM in 44 residences in Seattle (WA, US). In a recent study, Bi et al. [30] estimated outdoor infiltration and indoor contribution of $PM_{2.5}$ for 41 residential houses and 50 public buildings in California, US based on 91 PurpleAir indoor/outdoor monitor pairs. It found that site-specific proportions of indoor-generated $PM_{2.5}$ had a mean of 39.9% and a median of 41.9%, suggesting a relative substantial contribution of indoor sources to indoor air quality.

4.3. Limitations and conclusions

There are some limitations associated with the sampling campaign and modelling approach: 1) Co-location data were only compared to outdoor reference instrument data; therefore, there is uncertainty when outdoor correction factors were applied to the indoor instruments; 2) Outdoor measurements depended on where the sensors were situated (front or back door of the building), in particular when that building is near a local source (i.e. traffic emissions) [52]. The uncertainty in measurements from non-reference grade air quality sensors leads to uncertainty in estimation of infiltration factors and loss rates of air pollutants; 3) a mass balance model is unable to obtain unique pairs of infiltration factors and loss rates for non-indoor source periods (as shown in Fig. S3); and 4) uncertainty in a mass balance model exists due to assumptions of well-mixed air in a house and fixed infiltration, loss rate and air exchange rates during a time period.

Despite the above limitations, this study provided a comprehensive understanding of outdoor and indoor air quality in London homes. This work also examined air exchange rates between different homes and seasons. Key parameters for indoor air quality modelling including infiltration factors and loss rates of PM and NO_2 were investigated. A higher proportion of PM (F_{in}: 0.6–0.7) ingress into indoors from outdoor air was found than that for NO_2 (F_{in}: 0.42), suggesting loss rates of indoor pollutants is not only dependent on their deposition but other dynamic processes such as evaporation or chemical reactions. Ventilation plays a critical role in reducing indoor air pollution. Furthermore, we found a large contribution of indoor sources (26–37%) to indoor PM and NO_2 in London homes.

CRediT authorship contribution statement

Tuan V. Vu: Writing – review & editing, Writing – original draft, Visualization, Software, Methodology, Investigation. **Gregor B. Stewart:** Writing – review & editing, Writing – original draft, Visualization, Validation, Supervision, Methodology, Investigation, Conceptualization. **Nutthida Kitwiroon:** Validation, Software, Methodology, Formal analysis, Data curation. **Shanon Lim:** Writing – review & editing, Software, Methodology, Investigation, Formal analysis, Data curation. **Benjamin Barratt:** Writing – review & editing, Supervision, Resources, Investigation, Funding acquisition, Data curation. **Frank J. Kelly:** Writing – review & editing, Supervision, Resources, Investigation, Funding acquisition, Conceptualization. **Rhiannon Thompson:** Project administration, Formal analysis, Data curation. **Rachel B. Smith:** Writing – review & editing, Investigation, Formal analysis, Data curation. **Mireille B. Toledano:** Validation, Supervision, Resources, Project administration, Funding acquisition, Data curation. **Sean D. Beevers:** Writing – review & editing, Writing – original draft, Supervision, Resources, Methodology, Investigation, Funding acquisition, Conceptualization.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

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Appendix A. Supplementary data

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