Environment International 153 (2021) 106515

Contents lists available at ScienceDirect

Environment International

journal homepage: www.elsevier.com/locate/envint

The UK Integrated Assessment Model for source apportionment and air pollution policy applications to $PM_{2.5}$

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ARTICLE INFO

Handling Editor: Xavier Querol

Keywords:
Integrated assessment modelling
$PM_{2.5}$ concentrations
Population exposure
Exceedance of WHO guideline

ABSTRACT

Source apportionment and the effect of reducing individual sources is important input for the development of strategies to address air pollution. The UK Integrated Assessment Model, UKIAM, has been developed for this purpose as a flexible framework, combining information from different atmospheric dispersion models to cover different pollutant contributions, and span the range from European to local scale. In this paper we describe the UKIAM as developed for SO_2, NOx, NH_3, $PM_{2.5}$ and VOCs. We illustrate its versatility and application with assessment of current $PM_{2.5}$ concentrations and exposure of the UK population, as a case-study that has been used as the starting point to investigate potential improvement towards attainment of the WHO guideline of 10 μg/m^3.

1. Introduction

The UK Integrated Assessment Model (UKIAM) was originally developed as a tool to investigate abatement strategies for reducing UK emissions to comply with emission ceilings, set for the UK in the Gothenburg protocols to reduce transboundary air pollution, and the National Emission Ceilings Directive of the European Commission. An early version was described in Oxley et al. (2013). Since then the aim has been to establish a model that can represent current and future emission scenarios, and assess the effectiveness of abatement measures in reducing exposure of the UK population to air pollution and corresponding health benefits; and also the effects of pollution on natural ecosystems in the UK - in particular effects of nitrogen deposition on eutrophication and biodiversity. This development has taken place in parallel with international development of emission ceilings under the UNECE Air Convention, and integrated assessment modelling by IIASA with the GAINS model of European scale emission abatement scenarios (see description on IIASA web-site of the GAINS project, Amann et al.). It needed to cover the same pollutants SO_2, NOx, NH_3, $PM_{2.5}$ and VOCs that are subject to emission ceilings. Use of the model has since expanded to more detailed assessments of exposure of the UK population and exceedance of WHO guidelines for $PM_{2.5}$; also to investigate specific sources such as shipping; and to undertake more detailed assessment of future protection of natural habitats. The benefits of abatement strategies can then be set against the costs, setting national action within the international context.

The current version of UKIAM produces mapped concentrations on a 1 × 1 km grid over the UK of $PM_{2.5}$, including both primary and secondary contributions superimposed on background contributions; and of NOx and NO_2 concentrations. These are combined with population data to indicate population exposure across the UK or in selected areas or cities, which can be used to assess monetised health impacts, together with detailed source apportionment as illustrated below. Deposition of sulphur and nitrogen is also calculated on a 5 × 5 km grid in order to compare with critical loads for the protection of natural ecosystems. In this paper we shall illustrate the application of the model to $PM_{2.5}$ concentrations and exposure.

The model needs to be fast to run in order to analyse a wide range of emission scenarios quickly and undertake sensitivity studies including investigation of specific sources or contributions. It currently runs on a laptop in about 30 min, and produces a very large amount of data for subsequent interrogation on source apportionment and production of maps.

In order to cover contributions imported from other countries and from shipping, as well as from sources across the UK down to local contributions from traffic and urban sources, UKIAM uses source

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https://doi.org/10.1016/j.envint.2021.106515
Received 16 October 2020; Received in revised form 3 March 2021; Accepted 8 March 2021
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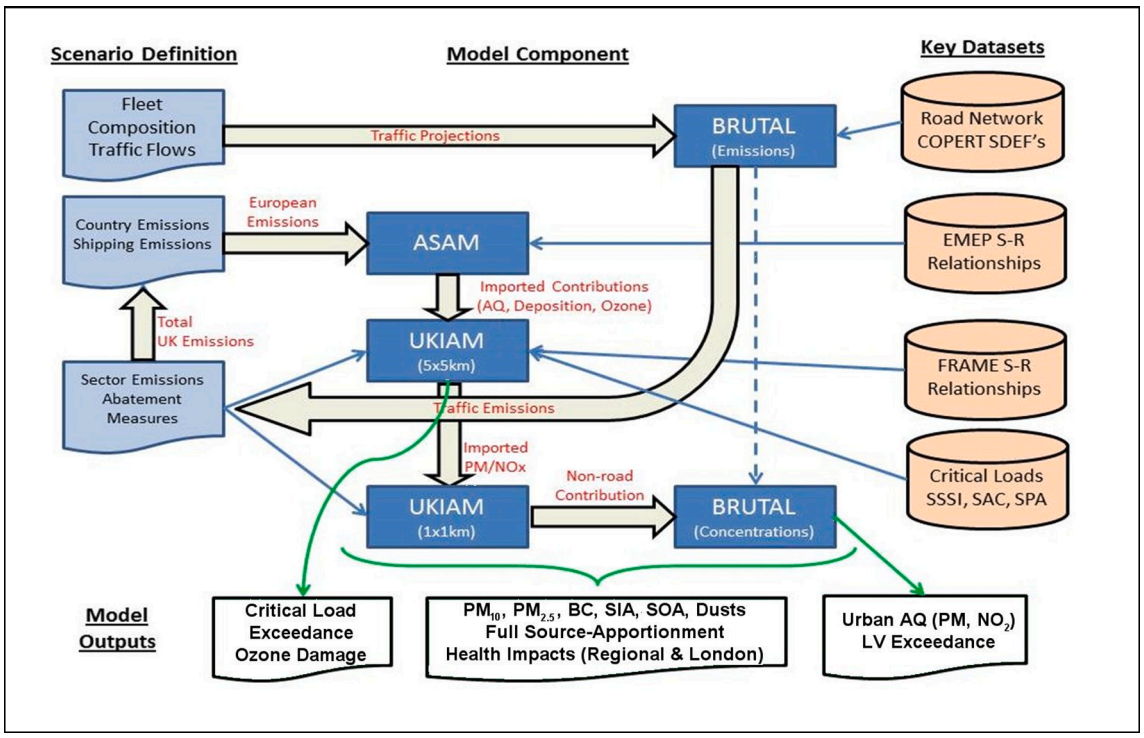


Fig. 1. Structure of the UKIAM Framework.

footprints derived by a range of atmospheric dispersion models covering different scales. This is described in more detail below, together with illustrations of source apportionment, and the situation in 2016 as the starting point for subsequent application of the model to investigate potential future progress towards achieving the WHO guideline of 10 $\mu g/m^3$ (see ApSimon et al. 2019a).

It is important that policy makers are suitably receptive to tools produced for the purposes of developing future air pollution control strategies, such as described in this paper, including consensus regarding the effectiveness and costs of pollution abatement measures. As such, this work is underpinned by emissions projections quantified by the National Atmospheric Emissions Inventory (NAEI) combined with strategies which capture the effectiveness, applicability and cost of alternative abatement measures. Suitable measures have been quantified in the Multi-Pollutant Measures Database (MPMD) to ensure consistency and confidence in measures applied in the UKIAM (see AMEC (2012) for further details).

2. Model description

In this section the current structure of the UKIAM framework is described, including the underlying atmospheric dispersion models that provide footprints of concentrations and deposition for different sources which are then adjusted in response to changes in emissions, and superimposed to map exposure across the UK. The structure is flexible, with the potential to substitute alternative emissions data or dispersion modelling for selected sources, or to add or remove sources. It is useful to highlight that the UKIAM is a framework. The *UKIAM Framework* consists of four separate modules: UKIAM1, operating at 1 × 1 km resolution (modelling primary PM and NOx/NO_2); UKIAM5, operating at 5 × 5 km resolution (secondary aerosols, and nitrogen and sulphur deposition); the BRUTAL model, capturing road transport; and the ASAM model, capturing transboundary contributions. Fig. 1 shows the overall model framework and the different components covering imported contributions from other countries and international shipping, and from UK sources where road traffic is treated in detail with the BRUTAL sub-model of the UK road network (Oxley et al. 2009).

The separate modules, covering different distance scales from the European scale down to the road-side, resolve source contributions to annual average concentrations on a 1 × 1 km grid, and deposition on a 5 × 5 km grid, spanning the UK and nearby sea areas. The ASAM module (ApSimon et al. 1994) covers the contributions from other countries and sea areas using the same atmospheric modelling of their individual footprints as in the GAINS model, based on the Eulerian EMEP model (Simpson et al. 2012). The responses to changes in emissions were derived by examining the effect of reducing individual pollutants from each country or sea area one at a time and examining the effect on concentrations and deposition across Europe. The resulting changes in concentration or deposition across the UK can then be normalised to provide source-receptor matrices reflecting the response to unit changes in emissions of each pollutant from each country or sea area.

UK emissions are based on the National Atmospheric Emissions Inventory, NAEI (https://naei.beis.gov.uk/), and future projections, with adjustments for abatement to investigate future scenarios towards attainment of national emission ceilings and improvement of air pollution in the UK. UKIAM distinguishes around 90 different sources as subdivisions of CORINAIR SNAP sectors (which distinguish emissions in eleven categories covering power generation, domestic and industrial combustion, industrial processes, solvents, transport, agriculture and natural emissions – see European Environment Agency); mapped on a 1 × 1 km grid by region (London, rest of England, Wales, Scotland and Northern Ireland). Emissions are assumed to be released at different (effective) stack heights which capture both an estimated chimney/release height plus plume rise; for example, road transport sources or domestic combustion (gas) assume emissions are released at ground level, whereas industrial sources may assume a stack height of 100–200 m, and power stations 300 m. Also modelled in more detail are contributions from UK and international shipping in the nearby sea areas surrounding the UK. For all these sources in the UK map area we use source-receptor matrices derived using the FRAME model for contributions to SO_4, NO_3 and NH_4 aerosol; and also for deposition of sulphur and nitrogen. These are derived in an analogous way to source-receptor

matrices from the EMEP model for the transboundary contributions. The FRAME model (Dore et al. 2007) is a Lagrangian model which is fast to run, and able to produce the large number of source-receptor matrices needed for the large number of different point and distributed sources in London, the rest of England, Wales, Scotland and Northern Ireland.

FRAME is based on an annual average wind-rose, and was developed at UKCEH (UK Centre for Ecology & Hydrology) with a focus on ammonia for which it has high resolution of surface layers to represent the competition between vertical diffusion and chemical transformation with deposition. It has been used to assess acid and nitrogen deposition in the UK, and critical load exceedance (Dore et al. 2007, 2012). FRAME has also been used to investigate spatially targeted ammonia mitigation measures and future changes in biodiversity. The performance of the FRAME model has been rigorously tested by a comparison with measurements of nitrogen and sulphur concentrations in precipitation, gas and particulate phase as part of a Defra (Department of Environment Food and Rural Affairs) model inter-comparison exercise, and was found to be fit for purpose (Dore et al. 2015).

For more detailed local scale modelling of NOx and primary PM, we use a Gaussian model, PPM, with adjustments for populated urban areas and topography. This is able to characterise the different types of stationary point and area/volume sources distinguished in UKIAM, allowing for different release heights and effects of buildings. This treatment of stationary sources is linked to the BRUTAL model (Oxley et al. 2009) which superimposes the contributions from traffic for primary PM, including non-exhaust emissions; and for NOx and primary NO_2, with a simple treatment of NOx and ozone chemistry when combined with non-traffic contributions (Oxley et al. 2009; 2013). Individual roads are superimposed on background concentrations to characterise road-side concentrations, with enhancement factors applied to concentrations to account for street canyon effects where it is assumed that street canyons become deeper, and thus more effectively trapping pollutants, as urban density increases, using population density as a proxy for urban density.

More details are provided in the illustration below of the application of UKIAM to assessment of $PM_{2.5}$ concentrations across the UK.

2.1. Application of UKIAM to $PM_{2.5}$ concentrations in the UK

The starting point for analysis of future scenarios towards reducing exposure to $PM_{2.5}$ and corresponding health impacts is establishing the current situation. In this section we illustrate the application of UKIAM to modelling concentrations of $PM_{2.5}$ across the UK in 2016 as a recent year for which detailed emissions data were available, and ascribing source apportionment.

2.2. Imported contributions from other countries

Some of the pollutants considered are typically transported in the atmosphere for up to a few days, with or without chemical transformation to secondary pollutants, before being re-deposited. This means that their range is European in scale, which is why emission ceilings have been set for each country in order to reduce transboundary air pollution in Europe. These have been negotiated under the Gothenburg protocols of the UNECE Air Convention, and set as binding targets for EC countries to achieve by 2030 under the National Emissions Ceilings Directive. These ceilings are prescribed individually for each country as % reductions to be achieved in emissions of each pollutant relative to emissions in 2005. They have been based on European scale modelling by IIASA with the GAINS model (Amann et al.), investigating technical abatement strategies that are aimed at maximising environmental benefits including health, and are cost effective. However, they do not reflect the more detailed information available within individual countries, and wider options and considerations in developing national policy.

As described above, the EMEP model of the Norwegian Meteorological Office (Simpson et al. 2012) has been used to represent atmospheric dispersion across Europe, and to provide source-receptor matrices representing the contribution of emissions from each country of each of the pollutants to gridded concentrations and deposition spanning Europe including the UK. These, together with similar data for different sea areas of Europe, have been used in the GAINS model: and made available for use in UKIAM to calculate what is imported from other countries. These data are incorporated in the ASAM module of UKIAM, which adjusts the contribution from each country or sea area according to the emission scenario. Emissions are based on reported emissions by each country for past and present years, and according to their emission ceilings for future target years of 2020, 2025 and 2030. For sea areas apart from those surrounding the UK as discussed below, the same assumptions have been made about current and future emissions as have been made by IIASA for the NECD development.

The transboundary (imported) contributions to PM and NOx concentrations in the UK which are calculated by the ASAM model include secondary inorganic aerosols (SIA) and primary PM and NOx; deposition of sulphur and nitrogen due to transboundary sources are also calculated. The summed imported contributions, excluding the UK and local sea areas, are mapped on to the finer UK grid, with some modification of wet deposition to allow for greater deposition over higher land, since smaller scale orographic effects are not represented in the coarser scale EMEP modelling. This is important in considering effects on ecosystems, although not the focus of this paper (Dore et al. 1992).

2.3. Modelling of pollution due to shipping in the nearby sea areas surrounding the UK

Of particular importance for the UK are shipping emissions in the nearby sea areas round the UK, including the North Sea, the very busy English Channel and the Irish Sea. Recent estimates of NOx emissions by Ricardo Energy and Environment (Ricardo 2017) based on Automatic Identification System (AIS) data from ships indicated 665kt of NOx emitted in these sea areas in 2016 from "UK international" (departing from or arriving in a UK port) and "in-transit" emissions from shipping passing the UK. This is in addition to 75kt of NOx from UK domestic shipping. These emissions encircling the UK compare with 870kt of NOx from total UK emissions in 2016, with a UK commitment for future reduction to 468kt by 2030 (a 73% reduction relative to UK emissions of 1735kt in 2005 for compliance with the National Emissions Ceiling Directive). Control of future shipping emissions in Europe under the International Maritime Organisation, IMO, has been much more limited than for land-based emissions, and is restricted to specific Emission Control Areas, ECAs in the North Sea and Baltic which only cover part of the shipping emissions round the UK. One priority for UKIAM has been to incorporate modelling of these shipping emissions in such a way that future scenarios for shipping, and their effect on air pollution in the UK could be investigated.

This includes the contribution of shipping to $PM_{2.5}$ concentrations and exposure across the UK, and deposition of nitrogen across the UK in relation to ecosystem protection, plus local effects on NO_2 concentrations due to emissions in port. To enable this the FRAME model of UKCEH has been used to provide source footprints of UK domestic and UK International plus in-transit shipping for emissions of SO_2 and NOx for secondary $PM_{2.5}$ and for nitrogen deposition. Emission projections to 2030 reflect growth rates in the 13 different vessel categories distinguished, combined with changes in emissions reflecting IMO legislation; and can be used to scale the current footprints in providing forecasts to 2030. Effects of shipping on NOx and NO_2 concentrations, and the small contribution of primary $PM_{2.5}$ emissions (dependent on Sulphur content of the fuels) are dominated by more local dispersion, using a simple Gaussian model based on annual average wind-rose consistent with the FRAME model.

A detailed contract report on the "Contribution of shipping emissions to pollutant concentrations and nitrogen deposition across the UK" has been produced for Defra based on this modelling with UKIAM (ApSimon

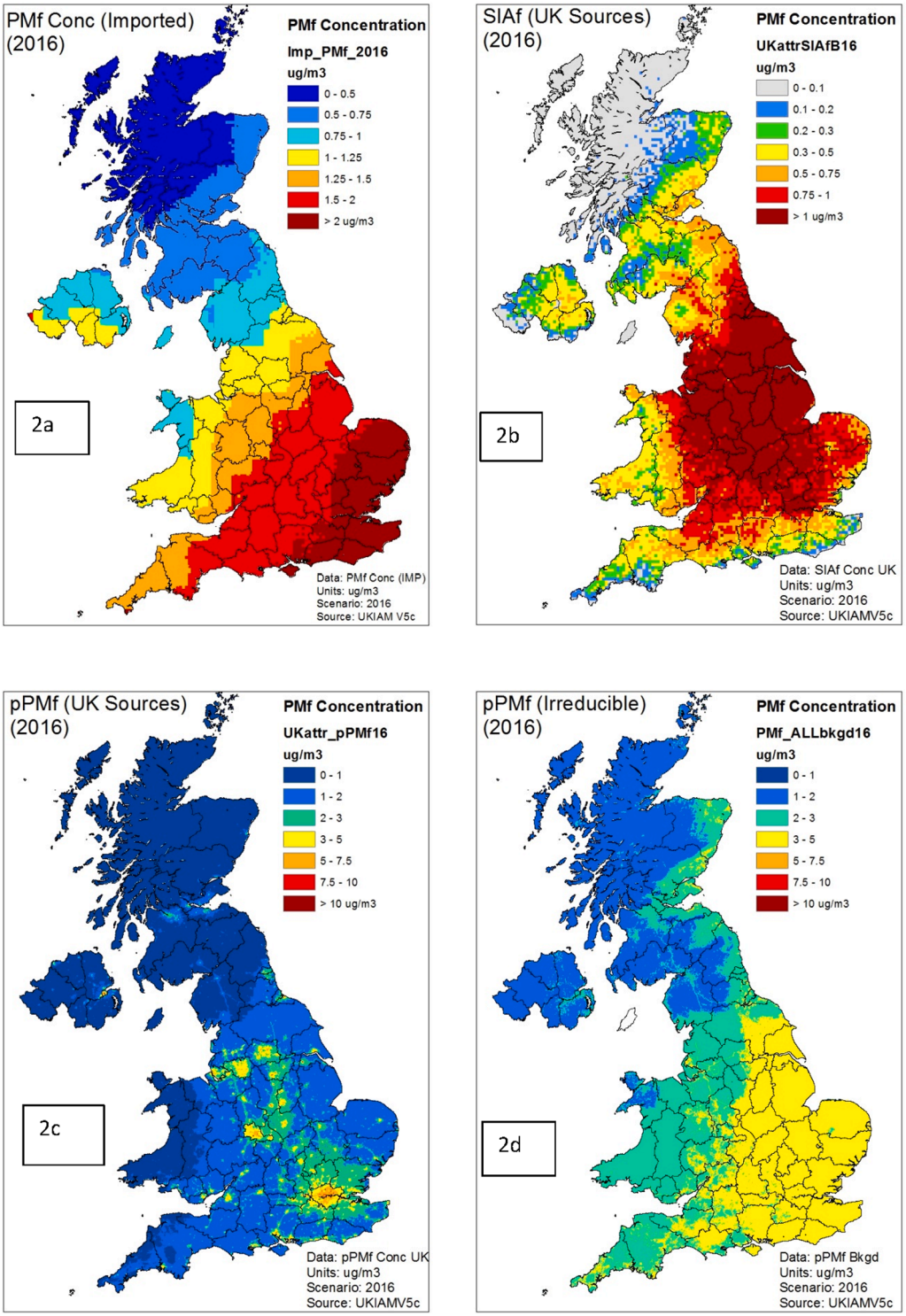


Fig. 2. Contributions to $PM_{2.5}$ concentrations in the UK, showing (a) transboundary (imported) contributions, including primary PM and secondary inorganic aerosols (SIA) from Europe and international shipping, (b) Secondary Inorganic Aerosols attributable to UK sources and national shipping, (c) Primary PM attributable to UK sources and national shipping, and (d) 'Irreducible' contributions which include natural dusts, sea salt, water and secondary organic aerosols.

et al. 2019b). This shows that the large NOx emissions from UK International plus in-transit shipping make a considerable contribution to $PM_{2.5}$ exposure in the UK, and health costs exceeding £1billion per year.

Fig. 2a shows the combined contribution to $PM_{2.5}$ concentrations across the UK in 2016 imported from other countries and from shipping.

2.4. Modelling the contribution of UK emissions

UKIAM currently distinguishes around 90 source categories in the UK as a mixture of point, area and volume sources with emissions of SO_2, NOx, NH_3, PM_{10}/$PM_{2.5}$, and VOCs mapped on to a 1 × 1 km grid covering England, Wales, Scotland, Northern Ireland and London. Emissions from the National Atmospheric Inventory, NAEI, are assigned to these 90 source categories in each region. These include individual

major point sources, for example a coal fired power station now converted to biomass, and arrays of smaller commercial or industrial sources or gas plants; and domestic sources such as wood-burning, gas, coal and oil which are treated separately to reflect different urban/rural distributions and emission characteristics. Agricultural sources of ammonia are broken down by animal category, plus fertiliser use, with additional non-agricultural emissions including anaerobic digestion.

Emissions from the different stationary sources (domestic, power, industrial, agricultural etc.) are specified in the UKIAM using the most recent NAEI emissions estimates together with projections to future years. This ensures consistency of representations with the official reporting of emissions in relation to international agreements such as the National Emissions Ceilings Directive (EU) and the Gothenburg Protocol (UN/ECE). The estimation of emissions captures both the spatial distribution of emissions from different sources (Tsagatakis et al. 2020) and specifies emissions relating to the Devolved Administrations (Scotland, Wales, Northern Ireland) (see Smith et al. 2020; Richmond et al. 2020).

Emissions from road traffic are treated differently, with the BRUTAL sub-model accumulating gridded emissions across the road network of the UK, using COPERT V5 emission factors combined with traffic flow data and the vehicle mix by type and Euro category (Oxley et al. 2009). Traffic flow data is mapped across the entire UK road network, based upon traffic counts at over 40,000 locations (https://roadtraffic.dft.gov.uk/), providing the composition of different vehicle types in the fleet (cars, LGV's, HGV's, buses etc.). These data are supplemented by fleet composition projections available from NAEI which specify the Euro technology split derived from DfT (Department for Transport) fleet turnover models (Pang & Murrells, 2019). Separate work has been undertaken on real-world emissions with a focus on diesel cars to compare with COPERT, and help explain a tendency to underestimate concentrations in congested urban conditions (O'Driscoll et al. 2016, 2018). Emissions are retained for the busiest roads in each grid square, based upon traffic flows, and used to calculate road-side concentrations which are superimposed on the background for the grid square.

To estimate concentrations and thus human exposure to $PM_{2.5}$, UKIAM calculates concentrations averaged over 1 × 1 km grid squares. This involves first estimating contributions from primary $PM_{2.5}$ sources dominated by local scale dispersion and modelled by the Gaussian PPM model. To this is added the contribution from UK sources and nearby shipping from secondary particulate SO_4, NO_3 and NH_4 aerosols, based on the FRAME model, and the contribution imported from other countries and more distant sea areas. As explained above, FRAME has been applied to examine the effect of reducing emissions of individual pollutants from each UKIAM source individually to see how this affects concentration and deposition across the UK, and the resulting source-receptor relationships incorporated in UKIAM. The FRAME model was chosen because, in addition to good performance in validation, the fast run-time made it possible to do the large number of runs for each pollutant from each of the approximately 90 sources in UKIAM in each region of the UK.

With respect to secondary inorganic aerosol the effect of reducing a precursor emission of one pollutant (SO_2, NOx or NH_3) on the combined mass of sulphate, nitrate and ammonium aerosol has to be estimated. The non-linear chemistry has implications for the linear scaling assumptions made in UKIAM to represent the effect of changing emissions; this is addressed in the following section.

For more local scale dispersion the PPM model is used for modelling primary $PM_{2.5}$ and NOx. This is a Gaussian model, and produces source footprints for point, area and volume sources. It uses an annual average wind-rose consistent with the FRAME modelling, with adjustments for rural to urban roughness and city environments, and for individual sources; for example to allow for release from chimneys for domestic heating with wood or coal, as compared with more variable discharge patterns over buildings from domestic gas. Domestic wood combustion is a very uncertain source (discussed further below) where the emissions have been estimated by Defra based upon assumptions about the proportion of wet/dry wood being burnt, and different spatial distributions of wood being burnt in boilers or on open fires.

Model performance has been tested against measurements (both background and roadside) and against other models (e.g. Stedman et al. 2007a), including a model inter-comparison study where all models under-predicted concentrations, which was attributed to under-estimation of real-world emissions (Carslaw et al. 2013). The performance of the UKIAM has been reviewed alongside other models by an expert group in the UK Atmospheric Dispersion Modelling Liaison Committee (https://admlc.com/), and was approved as 'fit for purpose'.

Fig. 2b and 2c show the contributions of UK sources in 2016 to fine secondary inorganic aerosol (based on scaling of FRAME source-receptor footprints), and to primary $PM_{2.5}$ concentrations. These are very different in character with the primary $PM_{2.5}$ concentrations showing sharp peaks in urban areas, especially in London. These local peaks are very important subsequently with regard to areas and populations exceeding the WHO guideline.

2.5. Non-linearities in modelling SIA

As noted above the non-linear chemistry of secondary inorganic aerosols has implications for the linear scaling assumptions made in UKIAM to represent the effect of changing emissions. Recognising the issue of non-linearity, and given the need to account for the bias implied by the 9% underestimation (see Appendix) of concentrations when estimating exposure or exceedance of the WHO guideline, investigations are ongoing to account for any losses of SIA concentrations due to either the non-linearities of the cross-pollutant chemistry or the effect of combining different models.

Initial comparisons of modelled SO_4, NO_3 and NH_4 concentrations with monitoring data from the UKEAP: Acid Gas & Aerosol Network (AGANET), suggest that a correction of 0.5–1 μg/m^3 in SIA concentrations may be appropriate and account for some of this bias. Comparisons with more complex Eulerian modelling also highlights inter-annual variations in meteorology where SIA concentrations in 2016 were relatively high. This work will be described in a future publication. Meanwhile in allowing for uncertainties we consider the sensitivity of results to a uniform under-prediction of 1 μg/m^3.

It is assumed that small changes to precursor emissions will not unduly impact on the non-linear chemistry. Independent checks suggest that, within the range of emission changes made in applying UKIAM, the effect is less important than other uncertainties and assumptions made, as discussed in the section on uncertainties below (Aleksankina et al. 2018).

2.6. Other contributions

The current WHO guideline of 10 μg/m^3 for annual $PM_{2.5}$ concentrations applies to the total mass of $PM_{2.5}$, and there are other sources contributing to total $PM_{2.5}$ concentrations, both secondary and primary, which need to be taken into account when estimating exceedance of the guideline. This is in addition to uncertainties in the contributions, and missing sources of primary $PM_{2.5}$ in the national emission inventory, as discussed below. Thus, there are additional contributions such as secondary organic aerosol (SOA) generated by biogenic emissions, and natural background dust; in order to capture this we have brought together data from other modelling studies to obtain a more complete picture. Currently we include SOA modelling as originally taken from the NAME model of the UK Met Office and revised and calibrated to match measurements. Having undertaken comparison with other modelling studies, these contributions will be revised when there is an improved understanding of anthropogenic contributions to SOA in addition to the major influence of biogenic VOCs. We are also aware of the role of IVOCs in contributing to $PM_{2.5}$ (VOCs of intermediate volatility) whose emissions are not yet quantified for inclusion in emission

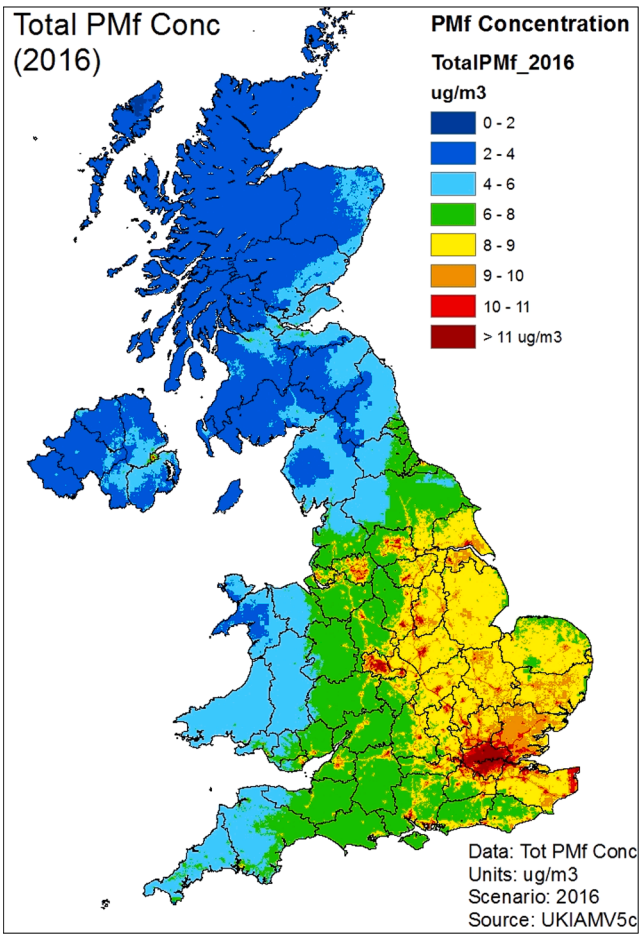


Fig. 3. Total $PM_{2.5}$ concentrations in 2016.

inventories (for example, see parallel modelling of $PM_{2.5}$ concentrations in the UK with the CMAQ model by Kitiwiroon et al. 2019).

Contributions of natural dust and sea salt have been provided by Ricardo as used in their Pollution Climate Model, PCM, which is used for regulatory purposes (Stedman et al. 2007b). We also add water as included in the EMEP modelling. Apart from some small reduction in water content with SIA concentrations these additional contributions are currently assumed to remain fixed when considering future scenarios, and when combined are referred to below as the "irreducible contribution" for which we do not have abatement options. The bottom right hand map in Fig. 2(d) shows our indicative estimate of this overall irreducible contribution, which is constructed by building up the contributions from these different natural sources, including SOA. Clearly there are large uncertainties and much needs to be done to improve the representation of these additional components. However, the map shows that there is a substantial combined contribution to $PM_{2.5}$ concentrations, amounting to over 3 μg/m^3 over large parts of England where the secondary inorganic aerosol is largest. Despite uncertainties it is important that this is taken into account when considering exceedance of the WHO guideline, and how much future abatement strategies can reduce this.

2.7. Population exposure

Fig. 3 brings all the separate contributions to $PM_{2.5}$ together, combining the imported contributions with UK contributions, superimposed on the irreducible contribution. These modelled concentrations have been compared with measurements (see Appendix), showing a tendency to underestimate by approximately 9%. Ongoing work on SIA concentrations reflects part of this under-prediction (see above), but other sources also contribute to the under-prediction such as sources not captured by the NAEI which include emissions from cooking or condensable particles, for example from woodburning.

These are discussed below, and together with inter-annual variability and other uncertainties, need to be taken into account when considering exceedance of the WHO guideline of 10 μg/m^3. The areas of the map in red and brown have modelled concentrations in the UK above 10 μg/m^3, and include an estimated 15 million people. Of these approximately 6 million people are within 1 μg/m^3 of the guideline, and within the range of model uncertainty. Correspondingly around 12.4 million people are in the orange area, only just below the guideline as modelled. And a further approximately 13.5 million people are in the yellow area between 1 and 2 μg/m^3 below the guideline, and could possibly be at risk of exceeding the guideline in a year with more extreme meteorology than 2016. It is clear that the peaks in primary $PM_{2.5}$ concentration in urban areas are closely related to areas of exceedance, with London as

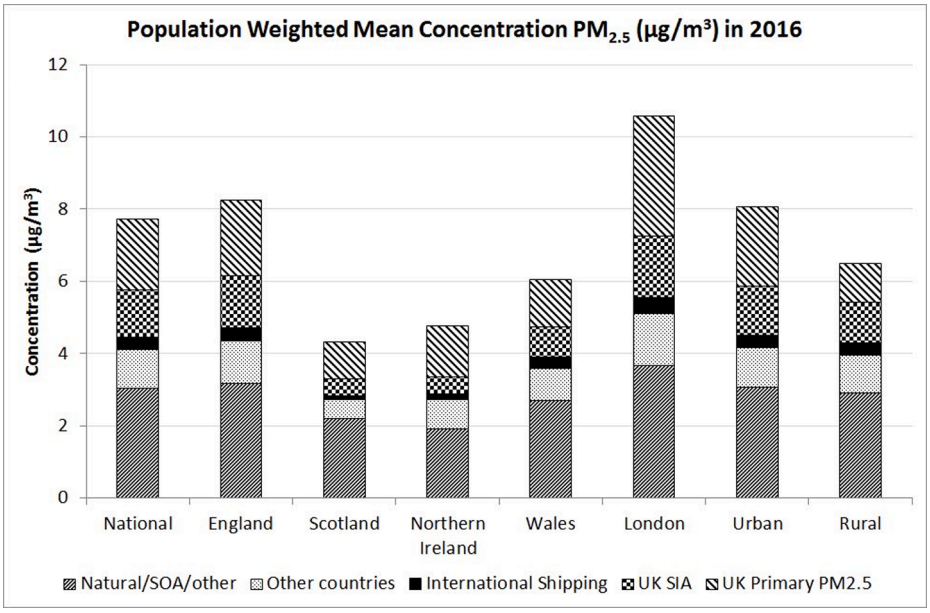


Fig. 4a. Population weighted mean concentrations by UK region in 2016.

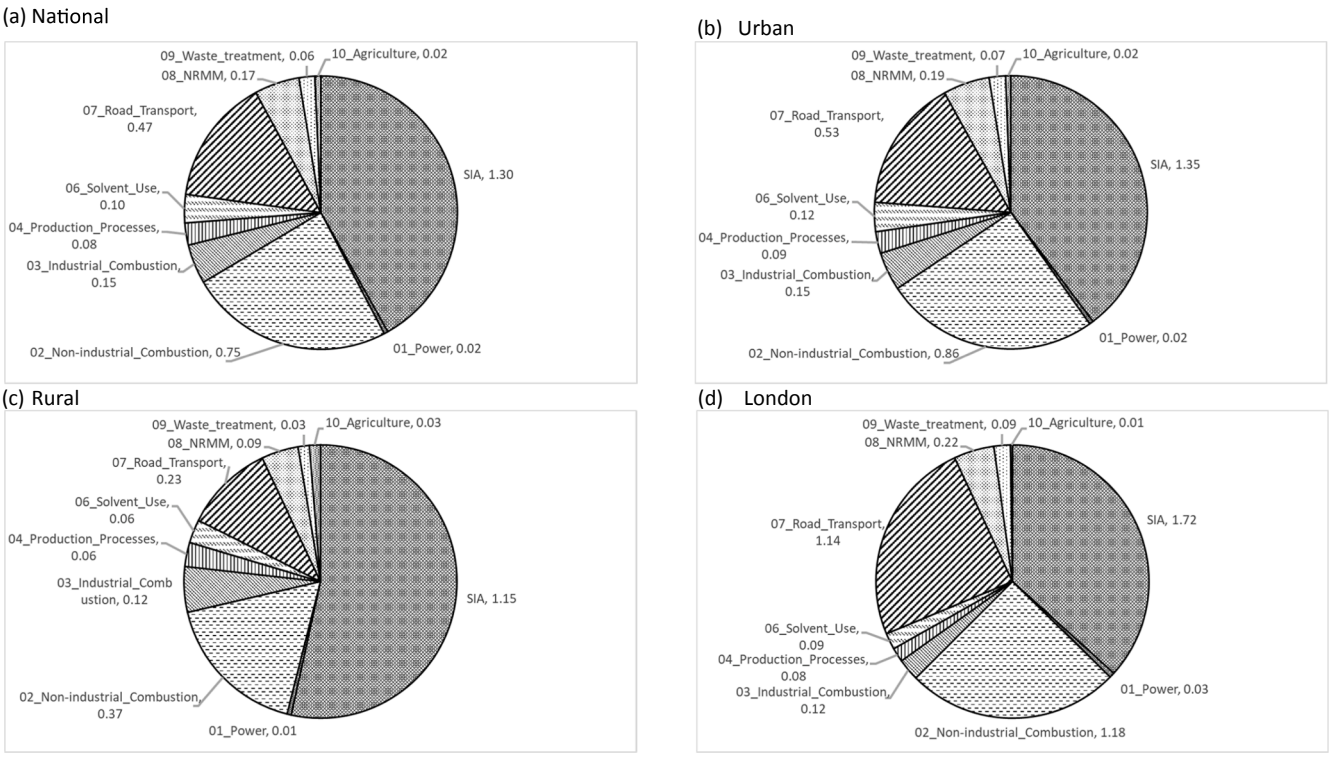


Fig. 4b. Source apportionment of UK contributions in 2016: to SIA, and primary PM by SNAP sector.

the biggest city situated in an area with a higher long-range background as an extreme case.

The next step is to assess the health effects on the UK population from this spatial distribution of $PM_{2.5}$. Combining the mapped $PM_{2.5}$ concentrations on a 1 × 1 km grid spanning the UK with population data gives an approximate estimate of population exposure, which can be used to assess health impacts. In order to compare different areas of the UK a useful indicator is the derived population weighted mean concentration, PWMC, obtained by dividing the population exposure for a given region by the population - that is to compare the average outdoor concentration to which people are exposed in different areas or regions.

$$PWMC = \sum_{ij} \cdot P_{ij} \times C_{ij} \Big/ \sum_{ij} \cdot P_{ij}$$

Where the summation is over grid cells i,j in the UK or sub-region with population P_{ij} and concentration C_{ij}.

For policy applications it is also useful to provide source-apportionment to give the relative importance of different sources, which is easily provided by the UKIAM modelling framework. The contributions of all sources captured by the UKIAM, to concentrations of primary and secondary air pollutants, and to deposition of SOx, NOx and NHx, are explicitly modelled individually, then combined to derive total concentrations and deposition. Source-receptor relationships are based on estimating the change in concentrations in a central year in the period under consideration (i.e. 2025); thus we minimise the magnitude of emissions changes and therefore non-linear effects.

This bottom-up approach avoids the need to disaggregate concentrations based on proxy (e.g. speciated) measurements and facilitates quantification of the contribution of individual sources alongside contributions from transboundary and shipping sources. Since this approach assumes linear responses it is important to note the limitations of this approach in relation to secondary pollutants which inherently reflect non-linear atmospheric chemistry (discussed above).

Fig. 4a provides an illustration of this source-apportionment, distinguishing urban and rural areas, and different regions of the UK and London as well as the total national picture. This reflects the geographical distribution shown in Fig. 3, with the highest PWMC for London due to higher long-range contributions to secondary inorganic aerosol, SIA (resulting from emissions in other countries, and from international shipping as well as UK emissions), as well as primary $PM_{2.5}$ from local emissions within the city. As expected, the population in urban areas has higher exposure than in rural areas, with a greater contribution from primary $PM_{2.5}$; and average exposure in England is higher than in other regions of the UK. Fig. 4 highlights an important message in that local measures can only address a small proportion of local PM concentrations. To address exceedance of the WHO guideline in urban areas a combination of local measures to reduce the peaks of primary concentration is needed alongside national and international efforts to reduce precursor emissions of SIA. However, a substantial proportion of $PM_{2.5}$ is attributable to non-anthropogenic sources not subject to controls.

As far as the UK contribution is concerned, which is under UK control, source-apportionment can be broken down in more detail as shown in Fig. 4b, where the combined contribution to SIA from UK emissions is shown alongside a sectoral breakdown of primary $PM_{2.5}$ contributions by SNAP sector. This can be used to pick out major sources, and to investigate uncertainties. As well as a major contribution from traffic where non-exhaust emissions are important, a particular example is wood burning in SNAP2 (non-industrial combustion) which is discussed in more detail below. Contributions to SIA from different sources have not been distinguished in Fig. 4b, but it should be noted that agricultural emissions of NH_3 have a significant effect on SIA concentrations, combining with NOx and SO_2 pre-cursor emissions from other sectors.

2.8. Exceedance of the WHO guideline

A particular application of UKIAM is to investigate exceedance of the current WHO guideline, where an initial goal was set for the UK to halve the number of people exceeding 10 $\mu g/m^3$ as an annual average (Defra Clean Air Strategy). The WHO guidance is based on epidemiological evidence related to total $PM_{2.5}$ by mass, which is why it has been

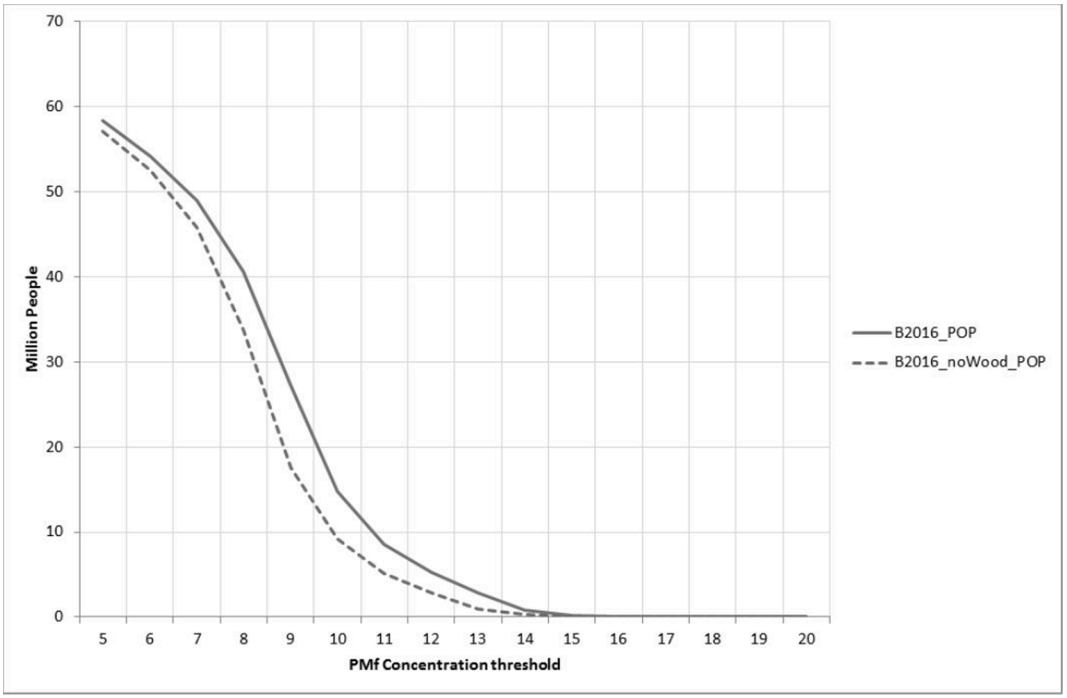


Fig. 5. Estimated distribution of population exposure in 2016. (millions of people exceeding concentration thresholds in μg/m^3).

Table 1
Modelled population (millions) exceeding threshold concentrations of $PM_{2.5}$.

Conc μg/m^3	National	England	Scotland	Northern Ireland	Wales	London	Urban	Rural
8	40.660	40.103	0.006	0.123	0.428	8.178	34.947	5.713
9	27.157	26.858	0.003	0.106	0.190	8.154	25.008	2.149
10	14.795	14.729	0.003	0.025	0.038	7.827	14.262	0.533
11	8.560	8.541	0.003	0.014	0.003	6.617	8.399	0.160

important to include the additional but uncertain contributions in the "irreducible" fraction. Fig. 5 and Table 1 show a graph of the estimated distribution of population exposure and the numbers of people exceeding 10 μg/m^3 in 2016, allowing for an error margin of plus 1 or minus 2 μg/m^3. It is clear that this error margin makes a large difference to the population above the guideline.

This is not unexpected as the maps show a large number of people close to the guideline, and small changes in the modelling estimates can make a big difference. As an example, an important but very uncertain source contributing to population exposure in the source apportionment above is domestic wood burning, where emissions are very dependent on the wood burnt and how it is used. The additional curve in Fig. 5 shows the effect of removing this source ('No-Wood') on the population exposure distribution. Emissions from wood burning will be sensitive to the moisture content of the wood burnt, and the population exposure will be very sensitive to both the assumed moisture content and the spatial distribution of emissions. Assumptions regarding wood burning are consistent with the NAEI, and potentially contribute up to 2.5 μg/m^3 to $PM_{2.5}$ concentrations in the south-east of the UK (AQEG 2017; Oxley & ApSimon, 2018).

Another disadvantage of setting a target based on reducing the number of people exceeding the WHO guideline is that it does not necessarily place emphasis on improvement for those exposed to the highest concentrations. Thus, it can be easier to meet such a target by making small improvements for those close to the guideline whilst making little change for the population in the areas with the highest concentrations. In London there is a hard core of the population experiencing concentrations well above the guideline whose numbers are unresponsive to increasingly ambitious abatement scenarios, but who are nevertheless benefiting from reductions in concentrations towards the guideline. For these people who are at the highest risk it is important to have an indicator that quantifies their reduction in exposure and associated health benefits.

The number of people exceeding the guideline is therefore not regarded as a reliable indicator for setting targets and assessing improvement.

Another way of looking at exceedance of the WHO guideline is to consider the population weighted mean exceedance, PWME, alongside the population weighted mean concentration, PWMC, described above. The UKIAM adds up the "accumulated exceedance" above any prescribed threshold concentration, t, by adding up the population in each grid cell above the threshold times any excess concentration; this is then divided by the summed population to get an average exceedance per person.

$$\mathrm{PWME} = \sum_{ij} \cdot \mathrm{P}_{ij} \times \max\left(\mathrm{C}_{ij} - \mathrm{t}; 0\right) \Big/ \sum_{ij} \cdot \mathrm{P}_{ij}$$

If the threshold "t" is set to zero this equates to the total population exposure, and dividing by the total population gives the population weighted mean concentration, PWMC, as defined above. This can be used directly in calculating health impacts as explained above. If the threshold is set to the WHO guideline then the UKIAM calculates the accumulated exceedance of the guideline, which on dividing by the population as above gives the population weighted mean exceedance, PWME. This is a much better indicator to show *improvement towards eliminating exceedance* of the guideline than the number of people above the guideline discussed above.

Table 2
Population Weighted Mean Exceedance of threshold concentrations (μg/m^3).

Threshold	National	England	Scotland	Northern Ireland	Wales	London	Urban	Rural
8	1.262	1.504	0.003	0.108	0.134	4.346	1.511	0.402
9	0.716	0.855	0.002	0.045	0.045	3.336	0.889	0.119
10	0.391	0.468	0.001	0.013	0.011	2.342	0.493	0.037
11	0.211	0.253	0.001	0.005	0.006	1.437	0.268	0.016

In this paper we have used the current WHO guideline of 10 μg/m^3 to illustrate how those populations experiencing higher exposure can be given special attention. However, this is not a no-effect threshold below which health impacts do not occur (Chen and Hoek 2020), and is currently being reviewed by the WHO. In estimating health impacts we assume no threshold and follow the advice of the UK's Committee on Medical Effects of Air Pollution (COMEAP) and their recommended risk coefficients applied to overall concentrations and exposure. Thus, reduction of PWMC below 10 μg/m^3 will always be beneficial to health, and reduction of PWME is important in order to focus policy on reducing impacts on the worst exposed populations.

These population weighted indicators of exceedance make it easy to make direct comparison of levels of risk in different areas as illustrated in Table 2, and correspondingly to indicate future improvements and benefits of abatement scenarios. To allow for uncertainties, values of PWME are also calculated for thresholds 1 μg/m^3 above and 2 μg/m^3 below the 10 μg/m^3 threshold. It is clear that London has much higher exceedance than the rest of the country, and that exceedance of the WHO guideline is small in Wales, Scotland and Northern Ireland. The higher exceedance in urban areas is largely due to the urban peaks in primary $PM_{2.5}$ concentration, emphasizing the need to address primary $PM_{2.5}$ emissions in reducing exceedance of the WHO guideline.

While investigating potential future abatement scenarios it has been found that PWMC and PWME are far more stable indicators, and less sensitive to model uncertainties, than the number of people exceeding the WHO guideline (ApSimon et al. 2019a). Using them in combination, a reduction in PWMC indicates the health benefits of abatement strategies which can be monetised, and PWME sets a target for improvement in the more polluted areas. This contrasts with a focus on reducing the number of people above the guideline which can lead to small improvements for those close to the guideline, but with little or no emphasis on the most exposed. In application of the UKIAM to investigate future scenarios we now routinely use both parameters to assess the effectiveness and compare with costs of implementation.

2.9. Uncertainties

Clearly there are many uncertainties in the modelling described above, which ongoing work is addressing with emphasis on $PM_{2.5}$. This includes emissions where many of the important sources identified in the source apportionment, such as wood-burning and non-exhaust emissions, are highly uncertain. There are also significant sources such as cooking not yet included in the National Atmospheric Emission Inventory, NAEI, used to define the baseline UK emissions in UKIAM. There may be additional contributions from IVOCs with intermediate volatility which are still very much at the fundamental research stage, and the Task Force on Emission Inventories & Projections (TFEIP) is working to recommend a practical approach to the inclusion (or not) of condensables in PM inventories (https://www.tfeip-secretariat.org/). Such missing sources, reflecting uncertainty in the inventory, may contribute to a systemic bias as shown in the under-estimation of concentrations shown in the comparison of modelled concentrations and measurements in the Appendix, which is also influenced by weather and inter-annual variability.

There are also assumptions and uncertainties in the atmospheric modelling. The effects of non-linearity in the chemistry can be significant, and further inter-comparison is in progress with more complex Eulerian modelling of SIA (where we have not wanted to overestimate future improvements due to emission reductions). In urban areas there are also many uncertainties in the way pollution disperses in and between streets, which are difficult to resolve even with very detailed CFD models (e.g. Woodward et al. 2019). Within 1 × 1 km grid cells there will be local peaks not resolved by the model, and the micro-scale complexities of hot-spots would be a very serious difficulty if the WHO guideline was applied as a limit value in an analogous way to the limit value for NO_2. This is another reason for our dual emphasis on PWMC and PWME as indicators for reducing both human exposure and health impacts, and making improvements for those urban areas with higher exposure rather than focusing on hot-spots.

In applying to future scenarios additional uncertainties arise in emission projections, and the effectiveness of abatement strategies, both in the UK and for imported contributions. The break-down of concentrations into different contributions shown above indicates the relative importance of UK and imported contributions, and also draws attention to those additional components currently kept constant in the "irreducible fraction". This is a substantial and very uncertain contribution to total $PM_{2.5}$ concentrations by mass, and includes secondary organic aerosol.

3. Conclusions

In this paper we have described the UK Integrated Assessment Model, UKIAM, and have illustrated its application to concentrations of $PM_{2.5}$ in the UK in 2016. We have shown how the model provides detailed source-apportionment, and can be used to pick out the contribution of important individual sources, such as domestic wood-burning, both to concentrations and exposure of the population both nationally and in different areas of the UK.

Clearly the long-range imported contribution from other countries and shipping is important, with the Gothenburg protocols and the National Emissions Ceiling Directive, NECD, crucial in bringing down emissions across Europe. The contribution from shipping is of increasing concern relative to imported contributions from other countries, with limited abatement measures in restricted emission control areas of the North Sea. With regard to UK emissions we have distinguished the longer range secondary inorganic aerosol which varies more slowly spatially across the UK, from the more local primary $PM_{2.5}$ contributions which enhance concentrations in populated urban areas. In line with WHO guidance on using total mass of $PM_{2.5}$ from all sources in assessing total health impacts, we have also included an approximate assessment of other contributions to $PM_{2.5}$ concentrations including natural sources.

There are many uncertainties in such modelling which are the subject of further investigation, including missing sources which may contribute to the tendency to underestimate concentrations compared with measurements. Unfortunately, some of the most important sources such as wood-burning and non-exhaust emissions, are also the most uncertain; also cooking is an additional source we have investigated but is excluded from the UK National Atmospheric Emissions Inventory. Nevertheless, this modelling provides useful insight into source apportionment, and because of the quick run time of the UKIAM, it is very easy to run sensitivity studies and introduce improvements. For example, with respect to non-exhaust emissions from tyre and brake-wear, we updated UKIAM from using constant emission factors for each vehicle category to the Tier 2 methodology with emission factors depending on

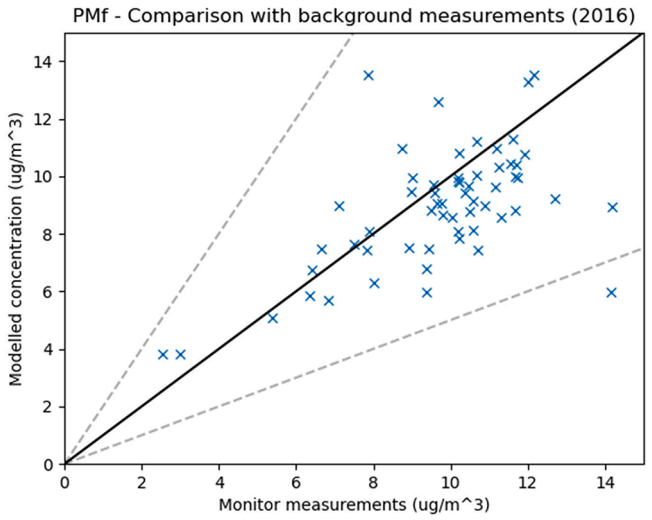


Fig. A1. Comparison of measured and modelled annual average background $PM_{2.5}$ concentrations for 2016. The dashed lines indicate the FAC2 limits and the full line indicates the line of equality.

Table A1
Comparison of model prediction and measurements for annual average background $PM_{2.5}$ concentrations in 2016. N = number of measurements, Obs. = mean of all measurements, Mod. = mean of all modelled concentrations, r = Pearson correlation, FAC2 = fraction of modelled values within a factor 2 of measured value, NMSE = Normalised Mean-Square Error, NAD = Normalised absolute difference.

N	Obs.	Mod.	r	FAC2	FB	NMSE	NAD
59	9.72	8.91	0.58	0.98	0.09	0.05	0.08

speed, and giving higher emissions in urban areas with more congestion and lower speeds. This introduced a significant difference in heavily trafficked urban areas coinciding with the areas of highest overall concentration.

Two indicators have been proposed as useful for setting targets for improvement. The first, *population weighted mean concentration, PWMC*, can be used directly in assessing overall exposure and health impacts, as well as in comparing exposure of different sub-groups of the population. The second, *population weighted mean exceedance, PWME*, can be used to provide a measure of the exceedance of the WHO guideline. Sensitivity to over or under prediction of modelled concentrations has been illustrated, but this is a far more robust indicator of exceedance than the number of the people exceeding such a threshold value for concentration.

In this paper we have focused on application of the model to the year 2016. This has been taken as a base year in applying the UKIAM to explore a range of future emission scenarios to 2030, when the NECD comes into force. This work has been published and can be found on the web-site of the UK Department of Environment, Food and Rural Affairs (ApSimon et al. 2019a).

Funding

This work has been supported by the Department of Environment, Food and Rural Affairs under grant ECM-53210:support for national air pollution control strategies.

CRediT authorship contribution statement

Helen ApSimon: Conceptualization, Methodology, Writing - original draft, Supervision, Project administration. **Tim Oxley:** Methodology, Writing - review & editing, Software, Validation, Investigation. **Huw Woodward:** Software, Data curation, Investigation. **Daniel Mehlig:** Software, Data curation. **Anthony Dore:** . **Mike Holland:** Methodology.

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.

Acknowledgement

This work reflects the personal views of the authors. Although the modelling work has been supported by the UK Department of Environment, Food and Rural Affairs, and has benefited from review by the UK Air Quality Expert Group and discussion with other colleagues, the findings and recommendations discussed here are those of the authors and do not necessarily represent the views of Defra or AQEG.

Appendix. Comparison of model results and measurements

The scatter plot below gives a comparison between modelled concentrations in 2016 as mapped in Fig. 3, above, and annual average measurements of $PM_{2.5}$ from the UK AURN monitoring network (https://uk-air.defra.gov.uk/networks). Table A1 shows performance measures calculated for these concentrations using 59 background measurement stations. The equations used to calculate the fractional bias (FB), Normalised Mean-Square Error (NMSE) and Normalised Absolute Difference (NAD) are shown below.

Some of the outliers coincide with significant differences across grid square boundaries, or have been explained by anomalies in mapping of emissions. Overall, there is a tendency to under-estimate (mean observed conc. = 9.72 μg/m^3, mean modelled conc. = 8.91 μg/m^3, FB = 9%), partly driven by the highest measured values which may be affected by local sources as compared with grid-average concentrations. This bias may also be in part due to non-linear effects as discussed in the paper and investigations are on-going regarding this issue. In making this comparison of measurements and modelling it should also be recognised that the modelling is based on annual average meteorology, and there could be significant inter-annual variability. Given these multiple sources of uncertainty, a good model performance is achieved, with 98% of modelled concentrations within a factor of 2 of the measured concentration.

Fractional bias:

$$FB = \frac{2\overline{C_o - C_p}}{\left(\overline{C_o} + \overline{C_p}\right)}$$

Normalised Mean-Square Error:

$$\mathrm{NMSE} = \frac{\overline{(C_o - C_p)^2}}{\left(\overline{C_o C_p}\right)}$$

Normalised Absolute Difference:

$$\mathrm{NAD} = \frac{\overline{|C_o - C_p|}}{\left(\overline{C_o} + \overline{C_p}\right)}$$

Here C_o is the measured, or observed, concentration and C_p is the modelled concentration. The overbar indicates that the mean is taken.

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Glossary

ADMS: ADMS is a pollution model for tackling air pollution problems in cities and towns, developed by Cambridge Environmental Research Consultants, http://www.cerc.co.uk/

AIS: Automatic Identification System, utilised to generate tracking information for ships

ASAM: Abatement Strategies Assessment Model [ApSimon et al., 1994], Imperial College London

BRUTAL: A road transport sub-model developed for the UKIAM [Oxley et al., 2009].

CMAQ: Community Multiscale Air Quality modelling system, http://www.cmaq-model.org/

COMEAP: Committee on the Medical Effects of Air Pollution, https://www.gov.uk/government/groups/committee-on-the-medical-effects-of-air-pollutants-comeap

COPERT: COPERT is the EU standard vehicle emissions calculator, available to download from https://www.emisia.com/utilities/copert/

Defra: Department of Environment, Food & Rural Affairs, http://www.defra.gov.uk/

EMEP: (1) Cooperative Programme for Monitoring and Evaluation of the Long-range Transmission of Air Pollutants in Europe (1984, Geneva Protocol) http://www.emep.int/ (2) Unified EMEP Eulerian model (Simpson et al., 2012)

FRAME: Fine Resolution Atmospheric Multi-species Exchange model [Fournier et al., 2004]

GAINS: Greenhouse gas and Air pollution INteractions and Synergies; a development of the RAINS model to address the inter-relationships with effects of greenhouse gasses (GHG), https://gains.iiasa.ac.at/models/

IIASA: International Institute for Applied Systems Analysis, Laxenburg, Austria http://www.iiasa.ac.at/

MPMD: Multi-Pollutant Measures Database (see AMEC 2012)

NAEI: National Atmospheric Emissions Inventory, https://naei.beis.gov.uk/

NECD: National Emissions Ceilings Directive (NECD) 2001/81/EC

$PM_{2.5}$: Airborne Particulate Matter less than 2.5 μm in diameter

PPM: (1) Primary Particulate Matter; (2) Primary Particulates Model [ApSimon et al., 2001]
PWMC: Population Weighted Mean concentration ($\mu g/m^3$) of an air pollutant, calculated as the sum of all exposures divided by the total population
PWME: Population Weighted Mean Exceedance, calculated as PWMC above a specified threshold
SIA: Secondary Inorganic Aerosols (ie. NH_4, SO_4 and NO_3)
SNAP: Selected Nomenclature for Air Pollution (https://www.eea.europa.eu/publications/EMEPCORINAIR/)
SOA: Secondary Organic Aerosols
SRM: Source-Receptor Matrices calculated by atmospheric dispersion models (eg. FRAME or EMEP) and used by integrated assessment models
TFEIP: UN/ECE Task Force on Emission Inventories & Projections https://www.tfeip-secretariat.org/
UKEAP: UK Eutrophying and Acidifying Pollutants network (http://uk-air.defra.gov.uk/networks/)
UKIAM: UK Integrated Assessment Model [Oxley et al., 2013; and described herein], Imperial College London
UNECE: United Nations / Economic Cooperation in Europe (UN/ECE), http://www.unece.org/
WHO: World Health Organisation, https://www.who.int/