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Reduced-form and complex ACTM modelling for air quality policy development: A model inter-comparison

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ABSTRACT

Simulation models can be valuable tools in supporting development of air pollution policy. However, exploration of future scenarios depends on reliable and robust modelling to provide confidence in outcomes which cannot be tested against measurements. Here we focus on the UK Integrated Assessment Model, a fast reduced-form model with a purpose to support policy development with modelling of multiple alternative future scenarios, and the EMEP4UK model which is a complex Eulerian Atmospheric Chemistry Transport Model requiring significant computing resources. The EMEP4UK model has been used to model selected core scenarios to compare with UKIAM, and to investigate sensitivity studies such as the interannual variability in response to meteorological differences between years. This model intercomparison addresses total $PM_{2.5}$, primary $PM_{2.5}$ and Secondary Inorganic Aerosol concentrations for a baseline of 2018 and selected scenarios for projections to 2040. This work has confirmed the robustness of the UK Integrated Assessment Model for assessing alternative futures through a direct comparison with EMEP4UK. Both models have shown good agreement with measurements, and EMEP4UK shows an ability to replicate past trends. These comparisons highlight how a combination of reduced-form modelling (UKIAM) and complex chemical transport modelling (EMEP4UK) can be effectively used in support of air pollution policy development, informing understanding of projected futures in the context of emerging evidence and uncertainties.

1. Introduction

Simulation models can be valuable tools in supporting development of air pollution policy, and different models can investigate a wide range of questions, which may be scientific, technical and/or socio-economic (McIntosh et al., 2007). Different models may have contrasting strengths and weaknesses and can often be distinguished as being policy models or research models, or identified as simple/complex, process-based or statistical; policy models may be simplified (reduced-form) representations but are very quick to run, and research models may be complex and evolving models capturing the latest scientific understanding. Models can be validated against measurements, although comparison with measurements can only be made for the current and historical situations; exploration of future scenarios depends on reliable and robust modelling to provide confidence in outcomes which cannot be tested against measurements.

Models should be evaluated on three levels. Firstly scientific, in that the model uses accepted, state-of-the-art representations; secondly operational, that it replicates observations adequately; and thirdly diagnostical, that it is fit-for-purpose and suitable for answering policy questions (see, Derwent et al., 2010). Both models discussed below are recognised as scientifically acceptable (Williams et al., 2011; AQEG, 2021), we show that they are consistent with observations, and we conclude that they are suitable for policy support. Model intercomparisons are often used to provide additional confidence in projected outcomes where observations are not yet available. Such intercomparisons are widespread in climate modelling (eg Duan et al., 2019), used for assessment of co-benefits of climate mitigation and air quality (Rao et al., 2016), or provide comparisons of multiple reduced complexity models (eg. Gilmore et al., 2019; Nicholls et al., 2020; Foley

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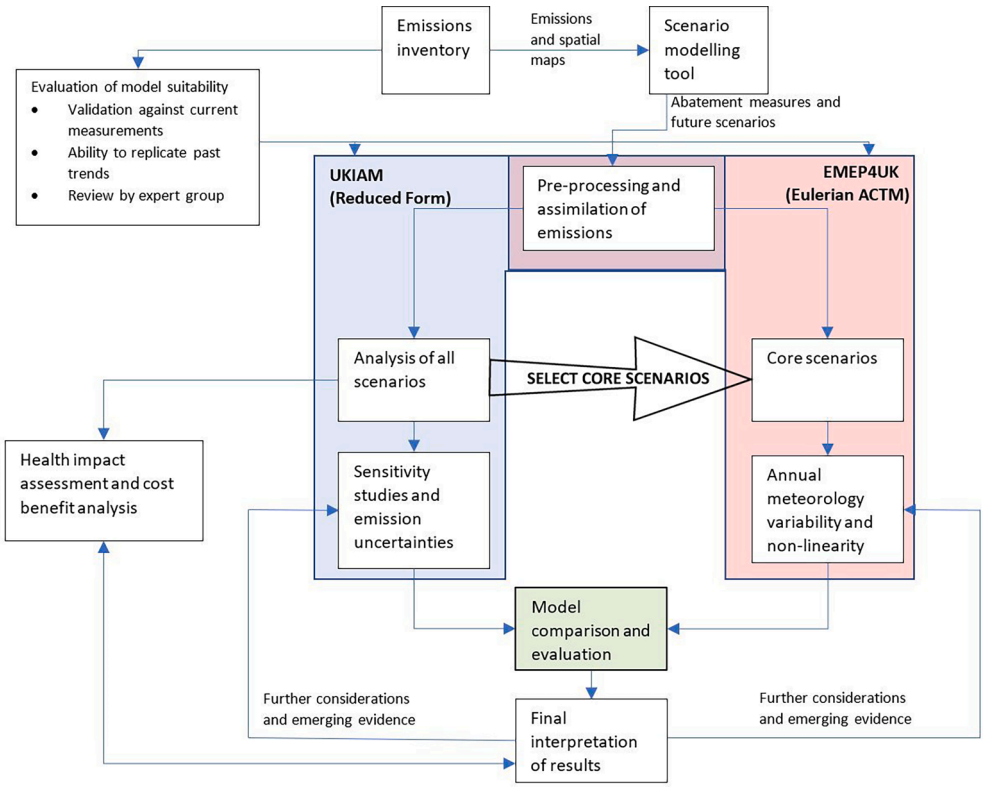

Fig. 1. Flow chart highlighting the broader context of policy development for which this model intercomparison provides confidence in the robustness of modelling projected futures; the shaded areas represent the focus of the work presented here.

et al., 2014). In contrast, Wu et al. (2021) compare a reduced-complexity model with a complex chemical transport model in China. For air quality modelling, model intercomparisons are less frequently undertaken (see, for example, Dore et al. (2015), Carslaw et al. (2013), Defra (2021)).

In this paper we focus on two models: The UK Integrated Assessment Model (UKIAM), which is a fast (reduced-form) model with a purpose to support policy development, and the EMEP4UK model which is a complex Eulerian Atmospheric Chemistry Transport Model (ACTM), which takes significant time and computing resources to run. Fig. 1 locates both models in the context of policy development, with the work presented here focussing on the shaded areas. Using the UKIAM, all potential emission reduction scenarios can be rapidly simulated, facilitating selection of a limited number of core scenarios for simulation using the more time and resource heavy EMEP4UK; this allows optimum use of models, firstly to filter out with a reduced form model those scenarios not meeting policy targets, and finally to model preferred scenarios in detail with the EMEP4UK ACTM. Detailed documentation of the models has been published elsewhere (eg. ApSimon et al., 2021a; Vieno et al., 2009), and extensive reporting of the policy scenarios has been made available for the policy consultation process (ApSimon et al., 2022).

Whereas the UKIAM is able to rapidly investigate many alternative policy scenarios, provide detailed source-apportionment of contributions, carry out sensitivity studies and sectoral analyses, apply source-specific abatement strategies, and quantify impacts on public health (see, for example, ApSimon et al., 2021a; 2022), the EMEP4UK model can address specific questions about the underlying atmospheric processes, such as the effects of inter-annual variations in meteorology, the effect of changing atmospheric composition resulting from significant reductions of precursor emissions or changing climate (ApSimon et al., 2022).

Models are widely applied to explore ex-ante what the impact of air pollution control strategies would be on ambient concentrations under a range of scenarios. Here, we document how both UKIAM and EMEP4UK have been utilised by the UK Department for Environment Food & Rural Affairs (Defra), which has the responsibility for setting air quality targets for reducing exposure to $PM_{2.5}$ in England as a key element of the forthcoming Environment Bill. We focus on $PM_{2.5}$ as the pollutant responsible for a large proportion of current adverse health impacts (RCP, 2016; Holgate, 2017). To reduce health impacts requires reductions in overall human exposure that are both ambitious and attainable, addressing those areas with the highest concentrations and greatest health impacts. Exploring the full scope of possible policy options involved modelling a wide range of potential future scenarios up to 2050, with different levels of ambition in abating emissions and the influence of climate measures. This paper briefly describes the modelling approach and the atmospheric models used in this work, and focusses on comparisons of selected scenarios between the UK Integrated Assessment Model (UKIAM) and the EMEP4UK model.

The analysis of a large number of scenarios required a model that is fast to run and that could represent total primary and secondary $PM_{2.5}$ concentrations spanning atmospheric transport over European to local scales, combining imported contributions from other countries and from international shipping, with more detailed consideration of UK emissions and enhanced concentrations in urban areas. Inevitably, there are many assumptions and uncertainties throughout the process, from quantification of emissions and atmospheric dispersion to impacts on health and the environment, which need to be recognised to inform robust policy decisions (see, for example, ApSimon et al., 2020; 2022). To safeguard that model assessments of the scenarios are able to account for variability in meteorological parameters and that potential non-linear responses to precursor emission changes are picked up, the more complex EMEP4UK model has been used to model selected scenarios to compare with UKIAM, and to investigate sensitivity studies such as the interannual variability in response to meteorological differences between years. This complements validation studies against measurements for UKIAM and provides greater confidence in policy

development where independent models show comparable outcomes for future scenarios.

The model intercomparison described here addresses total $PM_{2.5}$ concentrations, as well as specific aspects of primary $PM_{2.5}$ concentrations, and contributions of Secondary Inorganic Aerosols (SIA) to overall $PM_{2.5}$ concentration changes. This comparison includes a baseline for 2018 and selected scenarios for projections to 2040 to address changes in the anthropogenic contributions to air quality. Natural contributions (including Secondary Organic Aerosol (SOA)) are noted in discussions of uncertainty, but these require further investigation of representations which is beyond the scope of this paper.

2. Description of models

2.1. The UK Integrated Assessment Model

The UK Integrated Assessment Model, UKIAM, (ApSimon et al., 2021a; Oxley et al., 2013) has been developed as a scenario modelling tool and can run an individual scenario very rapidly once the required input data has been assembled. It was originally developed as a tool to investigate abatement strategies for reducing UK emissions to comply with national emission ceilings. These were set for the UK in the Gothenburg protocols to reduce transboundary air pollution, and in the National Emissions Ceilings Directive of the European Commission, now adopted in UK law. This required assessing the benefits of reducing emissions of SO_2, NO_x, NH_3, $PM_{2.5}$ and VOCs, both in the UK and the rest of Europe, both for human health and protection of ecosystems. UKIAM was originally developed to predict concentration changes, rather than absolute concentrations, but for this work simulates total $PM_{2.5}$ concentrations. Such calculation of total $PM_{2.5}$ introduces additional complexities with contributions from other sources (including uncertain natural sources, shipping and other transboundary contributions), together with the need to address the spatial variability in concentrations and exposure.

In UKIAM, UK emissions and future projections take, as the starting point, the National Atmospheric Emissions Inventory, NAEI, (Tsagatakis et al., 2019) and distinguish around 90 sources as subdivisions of CORINAIR (Core Inventory of Air Emissions) SNAP (Selected Nomenclature for Air Pollution) sectors. These define emissions in eleven categories, covering power generation, domestic and industrial combustion, industrial processes, solvents, transport and agricultural emissions. A sub-model, BRUTAL, simulates the road transport in more detail, accumulating emissions across different types of road on a bottom-up basis across the UK road network. See ApSimon et al., (2021a) and Oxley et al., (2009) for more detailed descriptions of the model.

The ASAM module (ApSimon et al., 1994) captures the imported $PM_{2.5}$ from other countries and sea areas, using the same atmospheric modelling of their individual contributions as in the GAINS model (http s://gains.iiasa.ac.at/models/), based on the European Eulerian EMEP model (Simpson et al., 2012). The responses of concentrations and deposition to changes in emissions were derived using source-receptor matrices reflecting the response to unit changes in emission of each pollutant from each country or sea area. The central focus here is the secondary inorganic aerosol, SIA, resulting from the emissions of NO_x, SO_2 and NH_3; with primary emissions giving only a small imported contribution.

Changes in SIA concentration due to UK sources are calculated in a similar way as the imported SIA, but use source-receptor matrices across the UK on a 5×5 km^2 grid for each source and pollutant as calculated by the FRAME model (Singles et al., 1998; Fournier et al., 2004; Dore et al., 2007). The Lagrangian FRAME model is fast to run and could undertake the large number of runs required to provide this substantial data set, also producing parallel data on deposition of sulphur and nitrogen required for assessing impacts on ecosystems. The FRAME model has also been applied to shipping in the seas surrounding the UK, where international shipping generates substantial NO_x emissions, contributing to SIA concentrations and nitrogen deposition across the UK (ApSimon et al., 2021b).

Using linear scaling of the above source footprints in accordance with changing emissions, provides a fast way of assessing changes in SIA contributions, but ignores the non-linear behaviour of chemical interactions between pollutants, and interactions with changes in the import from outside the UK. This is justified, providing the overall emission reductions do not change the chemical mix too far (Aleksankina et al., 2018; 2019), but does not provide an estimate of total concentrations starting from zero emissions. To overcome this, initial concentrations of SO_4^{2-}, NO_3^- and NH_4^+ components are matched to measurements for the current situation, taking an average of three years of data from the AGANET measurement network (to allow for interannual variability) and adding an additional mapped contribution across the UK.

It should be noted that the formation of SIA from the precursor gas emissions takes time, with subsequent removal on a time scale of a few days involving atmospheric transport over continental scales. The resulting concentration map varies relatively smoothly without localised peaks close to major sources, for example within urban areas. In contrast, primary emissions of $PM_{2.5}$ can give rise to sharp localised peaks in concentration close to the source. In UKIAM this is modelled with the PPM Gaussian model producing concentrations on a 1×1 km^2 grid, to match the resolution of the emissions data, and based on annual average wind-rose data. Adjustments are made to reflect source characteristics such as effective release height, and urban effects on dispersion. This results in enhanced urban concentrations and contributes to the higher overall exposure of urban populations to $PM_{2.5}$, especially in the extended city area of London.

There are other sources contributing to total $PM_{2.5}$ concentrations, both secondary and primary, which need to be considered when calculating total concentrations. These include secondary organic aerosol (SOA), as taken from the NAME model of the UK Met Office (Redington & Derwent, 2013) and calibrated to match measurements. Contributions from 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., 2007); and 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.

2.2. EMEP4UK

EMEP4UK is a full Eulerian atmospheric chemistry and transport model which simulates the emissions, transport, chemical transformations and deposition of a wide range of pollutants and provides hourly outputs (Vieno et al., 2009; Vieno et al., 2010; Vieno et al., 2014; Ots et al., 2016; Vieno et al., 2016a; Vieno et al., 2016b; Ots et al., 2018; Aleksankina et al., 2019; Carnell et al., 2019). It is a UK high-spatial resolution implementation of the European EMEP MSC-W model (Simpson et al., 2012; https://github.com/metno/emep-ctm), which is used within the framework of the UNECE Convention on Long-range Transboundary Air Pollution (CLRTAP) to assess country-to-country transport of air pollutants, the exceedance of critical loads thresholds for ecosystems and underpins the setting of European emission ceilings. The model simulates the various processes more mechanistically, whilst still with a simplicity that makes it applicable for multi-year, full-country simulations.

For the simulations here, EMEP4UK was based on EMEP model version rv4.36 and run at a resolution of about 3×3 km^2 over the British Isles, nested within a European domain with a horizontal resolution of 27×27 km^2. Fixed boundary concentrations were prescribed for the perimeter of the European domain, independent of scenario or year. The meteorological input data was generated with the Weather Research Forecast (WRF) model version 4.2.2 (Skamarock et al., 2008; https:

Table 1
Key statistical measures showing good agreement of both models with measurements for total $PM_{2.5}$. (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, RMSE = Root Mean-Square Error, NAD = Normalised absolute difference.).

Model	N	Obs	Mod	r	FAC2	FB	NMSE	RMSE	NAD
UKIAM	51	9.93	9.41	0.6	1	−0.05	0.03	1.7	0.07
EMEP4UK	51	9.93	9.43	0.61	1	−0.05	0.03	1.58	0.06

//www.wrf-model.org) which included data assimilation (Newtonian nudging) of the coarse-scale numerical weather prediction (NWP) model meteorological reanalysis with the US National Center for Environmental Prediction (NCEP) / National Center for Atmospheric Research (NCAR) Global Forecast System (GFS) at 1° resolution, every 6 h (Saha et al., 2014).

Compared with UKIAM, the EMEP4UK-WRF system is much more mechanistic and meteorologically explicit (see Table S.1, Supplementary Information). It therefore provides additional insights into the performance and robustness of the UKIAM and its use to support robust policy development. In particular, it provides:

(1) An assessment of whether UKIAM reasonably reflects the non-linear response of secondary inorganic aerosol (SIA) components to changes in precursor emissions, especially in the more extreme emission reduction scenarios.
(2) An indication of the change expected in the secondary organic aerosol (SOA) component, formed from biogenic and anthropogenic volatile organic compounds. This is kept constant in the UKIAM and therefore does not capture its response to emission changes.
(3) A quantification of the additional $PM_{2.5}$ that may be expected in years with particularly unfavorable meteorology.

The EMEP4UK-WRF modelling system has been tested widely against measurement data (Vieno et al., 2009; Vieno et al., 2010; Vieno et al., 2014; Ots et al., 2016; Vieno et al., 2016a; Vieno et al., 2016b; Ots et al., 2018; Aleksankina et al., 2019; Carnell et al., 2019) and shows good performance, except for roadside sites where the 3 km resolution is inadequate to capture the local enhancement. The UKIAM model, run at 1 km resolution, would be expected to perform better, although it will still not capture the true roadside increment. The EMEP4UK-WRF modelling system was run for the meteorological year of 2018 (to match the year of the baseline emissions) and, for comparison, 2003. The year of 2003 was selected because the meteorology led to higher-than-usual concentrations in $PM_{2.5}$ as can, for example, be seen in the peaks in the measured and modelled time-series of SO_4^{2-} and especially of NO_3^- (Fig. S.1a & S.1c). Vieno et al. (2014) analysed the reason for the elevated concentrations and showed that these were linked to extended periods of enhanced transport from continental Europe during February to April, which coincided with cool temperatures to favour the formation of ammonium nitrate (NH_4NO_3). There is no reason to believe that 2003 was an entirely unusual year, and whilst high ammonium nitrate episodes tend to dominate regional high $PM_{2.5}$ events in the UK (Yin and Harrison, 2008), other sources such as wildfires and volcano eruptions could also lead to elevated concentrations in particular years. The relative importance of ammonium nitrate could change in the future as its precursor emissions decrease, and other meteorological features could be more controlling for $PM_{2.5}$.

3. Emission scenarios

The scenarios we compare are based on emissions data provided by Defra using the Scenario Modelling Tool, SMT (https://smt.ricardo-aea.com/) to superimpose abatement measures on baseline NAEI projections, complemented by more detailed modelling of the road transport sector and electrification of the fleet. Modelling of fleet electrification includes a fleet turnover model to represent the evolution of the fleet up to 2050, with projections provided by DfT (Department for Transport), and is described elsewhere (Mehlig et al., 2021; ApSimon et al., 2022).

Many alternative scenarios have been run with the UKIAM to support the policy development process, based upon abatement strategies that could be described as of 'medium', 'high' or 'speculative' ambitions. The baseline (2018) and Business As Usual, High and Speculative scenarios for 2040 were also run using EMEP4UK, and these scenarios provide the basis of the model intercomparison reported here.

Abatement measures were developed following consultations in the form of sectoral workshops with stakeholders, quantifying potential emission reductions from different sources. The measures can relate to new technology, or to changes in behaviour. These have been superimposed on baseline (NAEI) emissions to give revised emissions using the SMT. This was specifically developed to enable the impact of different abatement measures on future emissions to be assessed. The tool works by applying specified measures to a baseline, modifying the emissions factor and/or activity level to produce a change in emissions. There are generally around 50–75 abatement measures defined for each scenario, with different implementation start dates, maximum uptake and profile of uptake over time.

The SMT only covers UK emissions. Imported contributions from other countries have been based on the "With Additional Measures" (WAM) scenario of IIASA (Amann et al., 2020). Emissions from shipping have been modelled based upon the Ricardo AIS tracking data for the domestic and international fleets around the coast of the UK and in the North and Irish Seas (ApSimon et al., 2021b).

The baseline scenario includes some adjustments to the original NAEI 2018 projections, reflecting various updates and revisions which include, for example, new emission factors for Euro6 RDE diesel cars, new regulations on domestic woodburning and red diesel and the impact of the Medium Combustion Plant Directive. The High scenarios include uptake of technologies considered likely to be implementable in the future, and an increased rate of behavioural change together with rapid uptake of measures over time. The Speculative scenario is the most ambitious and includes all feasible measures including emerging technologies and assumptions of significant behaviour change (ApSimon et al., 2022).

4. Results & discussion

To usefully compare the results of two distinct models such as UKIAM and EMEP4UK it is first necessary to report how each model compares with measurements (Section 4.1). This can of course only be carried out for current or past years, but it provides some confidence in each model so that results from projected future scenarios can be compared between models (Section 4.2).

It is important to re-emphasise some significant differences between the two models (see Table S.1). The UKIAM was originally developed to assess emission abatement strategies at a national scale, and models concentrations as annual averages. This means that seasonal variations in emissions (eg. agricultural NH_3) or meteorological episodes, are not well represented; this is reflected in the annual average source-receptor relationships calculated using the FRAME model. On the other hand, EMEP4UK-WRF is a complex ACTM which operates at much finer temporal resolution, with the ability to capture episodes and inter-annual

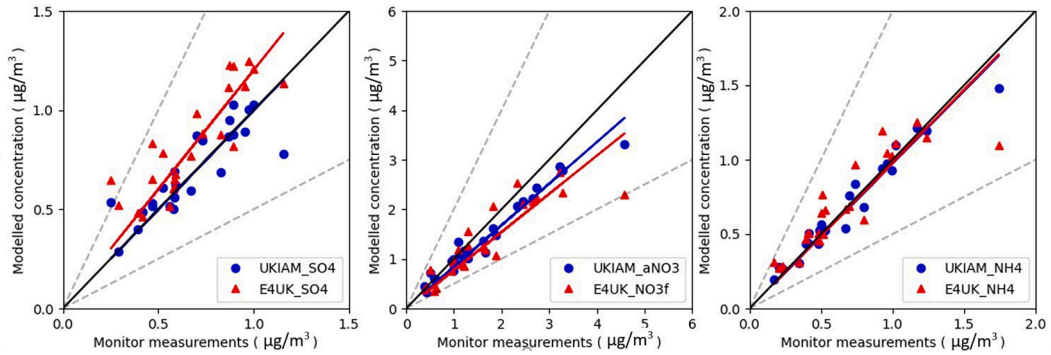

Fig. 2. Comparison of measured and modelled SIA concentrations for 2018. The blue/red lines are best fit lines through the origin. Dashed lines indicate the FAC2 limits & the full line indicates the line of equality. Panels display individual components of secondary inorganic aerosol, including sulphates (left), nitrates (centre) and ammonium (right). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

variability in meteorology.

A key objective of the comparisons is to highlight how the two models can best be deployed to provide robust modelling of projected futures for the development of air pollution policy (see Fig. 1). A large number of abatement scenarios can be quickly analysed using the reduced-form model, to then select a small number of core scenarios to investigate further with a full ACTM. The comparisons presented below are focussed in this context, and key statistical measures are reported (see Table 1). Conclusions are made in this context, and if the focus of observation changes, for example to a limited location, uncertainties and other model artefacts may need to be considered, and conclusions may become less certain (see Section 4.3).

4.1. Comparison with measurements

To demonstrate the skill in EMEP4UK-WRF to reflect concentrations and trends in SIA components over time, Fig. S.1 shows a comparison of modelled trends and site-specific model predictions against the measurements of the UK AGANET network. The model reproduces the measurements well both spatially and temporally, but there is a tendency for the model to underestimate the trend and to overestimate concentrations after 2010. This would indicate that EMEP4UK is, if anything, conservative in predicting the SIA reductions that may be achievable through reductions in precursor gas emissions (NH_3, NO_x, SO_2). This assessment is only indicative, however, as the model / measurement comparison heavily relies on the trend in the emissions to be correctly represented in the historic and current emissions inventories. Analysis of satellite observations, for example, has suggested that the NAEI may underestimate NH_3 emissions by 30 % (Marais et al., 2021), although the uncertainties in this independent approach are likely no smaller than in the NAEI itself. The impact of meteorology on agricultural emissions of NH_3 and soil emissions of NO_x is also not reflected in the NAEI and its trend (Sutton et al., 2013).

Although the work presented here reflects annual averages, the EMEP4UK model has been routinely evaluated against observations from the monthly UKEAP National Ammonia Monitoring Network (NAMN) for ammonia, the Acid Gas and Aerosol Network (AGANET) for aerosol inorganic chemical components and acid gases (https://www.pollutantdeposition.ceh.ac.uk/aganet), and the Automatic Urban and Rural Network (AURN) (https://uk-air.defra.gov.uk/networks/network-info?view=aurn) for hourly NOx, SO_2, $PM_{2.5}$, PM_{10}, and Ozone. Vieno et al. (2014) document comparison with the monthly NO_3^- and SO_4^{2-} surface concentrations at four AGANET selected sites for the years 2001–2010. The EMEP4UK model was able to capture the inter-annual variability as well as the monthly nitrate and sulphate. The hourly SIA components have also been compared with the MARGA (Monitor for AeRosols and Gases in Air) hourly data for the spring elevated $PM_{2.5}$ episode in 2014 (Vieno et al. 2016a). For 2018, modelled seasonal cycles

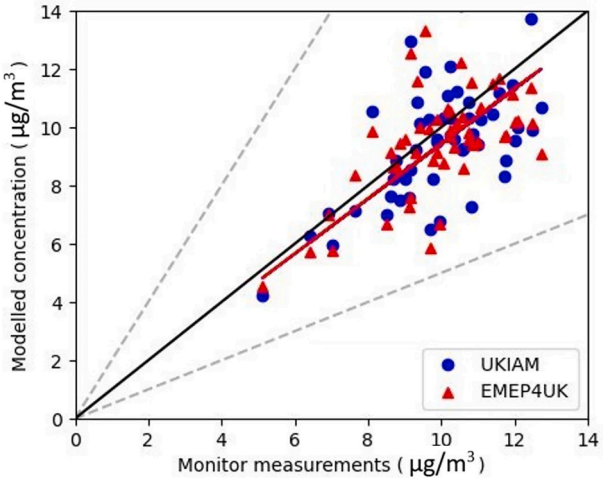

Fig. 3. Comparison of total $PM_{2.5}$ concentrations with observations for 2018 from the Automatic Urban and Rural Network (AURN) monitoring network.

track those averaged over the AGANET measurement sites closely, especially for NO_3^- and NH_4^+. Each month reflects an average over 11 to 23 sites, depending on data coverage of the month; see Fig. S.2 for details. Furthermore, the EMEP MSC-W model, from which EMEP4UK is derived, has been extensively evaluated with the results published by EMEP (https://aeroval.met.no/evaluation.php?project=emep-trends&exp_name=2005-2019&station=WORLD). EMEP-WRF has also been evaluated at the global scale for monthly NH_3 (Ge et al. 2021) in addition to other annual average observations. Given that the EMEP4UK-WRF model is virtually identical, the SIA scheme formation has been evaluated against observations across Europe.

For the Baseline 2018 scenario presented here, representing the current year for these analyses, the modelled concentrations of each SIA component are compared with measurements, showing good agreement (see Fig. 2). Table S.2 (Supplementary Information) provides a summary of key performance measures for the two models, showing that both models compare well against the measurements despite a degree of underestimation for the NO_3^-. Although the UKIAM is an annual model, a good comparison with EMEP4UK suggests we can infer that the annualised results of the UKIAM are not unduly affected by seasonal variations. The UKIAM cannot quantify seasonal variations itself, but used in combination with EMEP4UK (see Fig. 1) the potential effects of inter-annual variability in meteorology or non-linearity in SIA chemistry can be quantified alongside future scenarios modelled by the UKIAM.

Other contributions from SOA, natural dusts, sea salt and water content represent a substantial addition to total $PM_{2.5}$, amounting to over 3 µg m^{-3} for parts of England. Clearly, there are large uncertainties

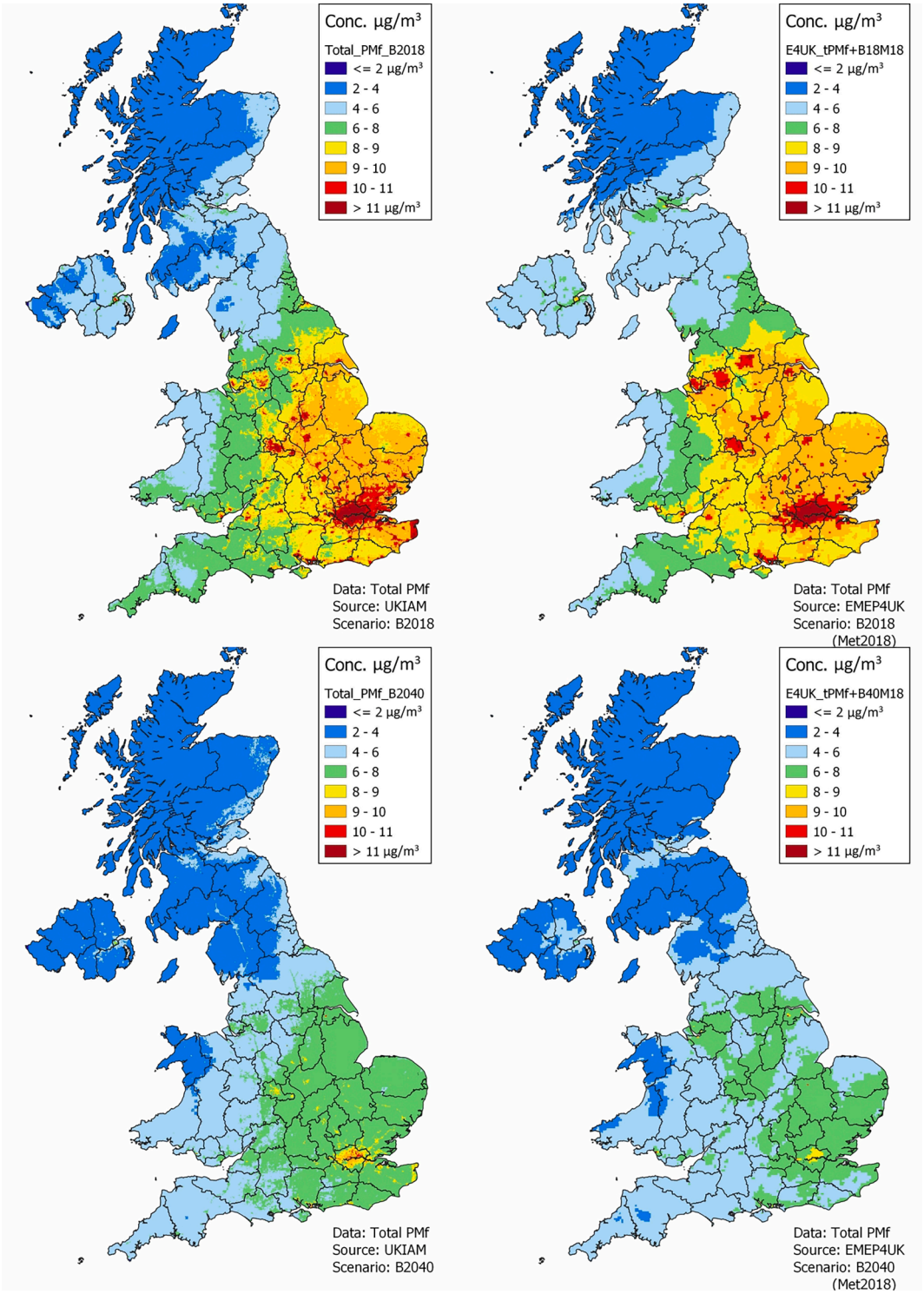

Fig. 4. Comparison of UKIAM results with EMEP4UK for 2018 emissions (upper) and for 2040 baseline emissions (lower).

(see ApSimon et al., 2020; 2022), but it is important that this contribution is considered when calculating total $PM_{2.5}$ concentrations.

With the contributions from all sources of PM accounted for, subject to caveats about uncertainties and sources missing from inventories (ApSimon et al., 2022), the modelled concentrations of total $PM_{2.5}$ from both UKIAM and EMEP4UK compare well against measurements of background $PM_{2.5}$ from the AURN monitoring network for 2018 (See Fig. 3). Table 1 presents the key statistical measures confirming this good agreement of both models with measurements.

4.2. Scenario comparison

In Table S.1 we document the differences between the models in relation to model setup and assumptions. For example, whereas UKIAM uses fixed annual average meteorology consistent with the FRAME modelling (Dore et al., 2007), EMEP4UK uses explicit meteorology's for specified years. The effect of meteorology is illustrated in Fig. 6 (below) in relation to modelled SIA concentrations for 2018 and 2040; modelled using different meteorological years, taking 2018 as an average year and

Table 2
Comparison of population weighted mean concentrations for Total $PM_{2.5}$.

PWMC in μg m^{-3}	National	London	Urban	Rural
2018				
UKIAM	9.2	12.3	9.6	7.8
EMEP4UK (2018 meteorology)	9.2	11.7	9.5	8.1
EMEP4UK (2003 meteorology)	10.1	12.7	10.5	9
2040				
UKIAM	6.8	9.4	7.1	5.8
EMEP4UK (2018 meteorology)	6.2	7.9	6.4	5.5
EMEP4UK (2003 meteorology)	6.7	8.3	6.9	5.9

2003 as a more adverse meteorological year. For the comparisons presented below, the same emission data has been used as far as possible, based on NAEI emissions for the UK, but broken down into the sources differentiated by UKIAM, and by SNAP sector for EMEP4UK. The same emissions have been used for other countries, but there are some variations in the shipping emissions, which are represented differently in the two models. We compare results for the baseline situation in 2018, and for a future scenario with much lower emissions in 2040.

Fig. 4 shows maps comparing total $PM_{2.5}$ concentrations from the two models for the baseline scenario in 2018 and 2040 respectively, with EMEP4UK using 2018 meteorology. The maps for 2018 show very similar concentrations, both corresponding to the same population weighted mean concentration of 9.2 μg m^{-3}, although if 2003 meteorology had been used with EMEP4UK the value would increase by almost one μg m^{-3} to 10.1 μg m^{-3}. (See Table 2).

In Fig. 4(c&d), with an equivalent comparison for 2040 using the same 2018 meteorology, both models show a large improvement compared to the base year 2018. However, the EMEP4UK concentrations of total $PM_{2.5}$ are overall a little lower than UKIAM, giving a population weighted mean concentration of 6.2 μg m^{-3} as compared with 6.8 μg m^{-3} from UKIAM. This implies that UKIAM tends to show a smaller improvement than EMEP4UK, which is what might be expected from the simplified, linear approximation along with a non-linear SIA adjustment in UKIAM. However, using the more severe 2003 meteorology in EMEP4UK gives higher concentrations equivalent to a population weighted mean concentration of 6.7 μg m^{-3}, almost the same as UKIAM. This suggests that the difference between UKIAM and EMEP4UK is within the general range of uncertainty due to variations in meteorology.

The next question is how the models compare with respect to individual components, in particular the primary $PM_{2.5}$ concentrations and the secondary inorganic aerosol, SIA, as the two components responding to the different emissions and their abatement. Fig. 5 shows a comparison of the primary $PM_{2.5}$ concentrations, as modelled by UKIAM and EMEP4UK for 2018 and 2040, where local emissions and urban areas generate a large spatial variability. These show that UKIAM tends to give slightly higher concentrations in urban areas, and slightly lower values in rural areas - which may be partially explained by the finer 1x1 km^2 grid resolution in UKIAM. Table 3 gives a comparison of PWMC values for 2018 showing close agreement between the two models.

The comparison of modelled SIA concentrations is particularly important because of the complex chemistry, and the simplified linear approach in UKIAM based on source-receptor matrices with a non-linearity adjustment as described above. Fig. 6 provides a comparison of total SIA for the baseline scenario in 2018 and 2040, respectively, with the EMEP4UK concentrations calculated using both the 2018 meteorology and the 2003 meteorology, the latter resulting in higher concentrations.

The maps show that, in 2018, the UKIAM SIA concentrations are close to the EMEP4UK concentrations using the 2018 meteorology, with a slightly lower population weighted mean concentration of 3.8 μg m^{-3} for UKIAM as compared with 4.1 μg m^{-3} for EMEP4UK. In 2040, when emissions are considerably lower, the UKIAM values have reduced less than with EMEP4UK (to 2.7 μg m^{-3} and 2.3 μg m^{-3}, respectively – see Table S.2), and are now closer to the EMEP4UK concentrations calculated for the less favourable meteorology of 2003. This is what was expected with the linear approximation used in UKIAM, and no change in the non-linear SIA adjustment. This means that UKIAM results for SIA reductions may be conservative, but they are still within the uncertainty range due to interannual variability in meteorology.

Fig. 7 shows comparison of the individual SIA components, NH_4^+, NO_3^- aerosol, and SO_4^{2-} (with the EMEP4UK concentrations based on 2018 meteorology), showing reasonable agreement between the models for individual SIA components (see also Table 4). This is also reflected in Fig. 2 where the SIA components from both models are compared with observations for 2018 from the AGANET monitoring network.

Further work is planned to compare other components, including SOA - where the calculations of concentrations with EMEP4UK show little change in the anthropogenic component (ApSimon et al., 2022). This is consistent with the assumption in UKIAM that SOA remains constant. But there are further questions, such as the role of IVOCs, which need to be addressed as the scientific understanding advances. Large uncertainties also arise in the contribution of other sources, including natural dusts. These other contributions are important, as together with the water content they add up to between 3 and 4 μg m^{-3} in some areas. They will be increasingly significant when considering the recently revised WHO guideline of 5 μg m^{-3} for annual average $PM_{2.5}$ concentrations (WHO, 2021).

Table 2 provides a summary of Population Weighted Mean Concentrations (PWMC) for total $PM_{2.5}$ calculated with UKIAM and EMEP4UK, also showing the effect of different meteorology in 2018 and 2003. The comparison is encouraging, with differences between the reduced-form UKIAM modelling based on annual average meteorology and the more sophisticated EMEP4UK modelling being generally within the interannual variability suggested by EMEP4UK. However, UKIAM tends to give higher peak concentrations in urban areas, reflecting the finer grid resolution of primary $PM_{2.5}$ contributions.

These comparisons are showing good agreement between the models for 2018, both in relation to observations (see Figs. 2 & 3 and Table 1) and direct comparison between the models for population weighted mean concentrations (see Tables 2, 3 & 4) and spatial variations (Figs. 4-7). Agreement between the models is supported by the grid-level comparisons shown in Fig. 8, which suggest that the EMEP4UK model is generally about 5 % higher than the UKIAM for both total $PM_{2.5}$ and SIA concentrations. Fig. 8(a) shows the comparison at monitoring locations, and Fig. 8(b) shows a heat plot of the comparison across all cells in the model domain. Note that multiple UKIAM cells may be compared with single EMEP4UK cells owing to the 1x1km^2 and 3x3km^2 resolution of the models, respectively. Some outliers may also be evident, for example due to differences in definition of the coastline at these resolutions, with SIA source-receptor relationships calculated at 5x5km^2 resolution, and concentrations provided for land-based cells. Statistical measures of the comparisons in Fig. 8 are provided in the Supplementary Information. Reductions in concentrations between 2018 and 2040 are less using the UKIAM, which is more conservative, with EMEP4UK showing concentrations between 11 % (total $PM_{2.5}$) and 19 % (SIA) lower than UKIAM (see Fig. S.5).

Two additional comparisons have also been carried out based upon emission scenarios for 2040 which reflect a High ambition scenario (H2040) and a Speculative scenario (S2040). As would be expected, both models show further reductions in concentrations. Maps of total $PM_{2.5}$ concentrations, and maps of SIA concentrations are provided in Supplementary Information, along with tables of population weighted mean concentrations. Across all the scenarios, the changes are consistent with expectations given the modelled responses to the reductions in concentrations between 2018 and 2040 shown above. Fig. 9 summarises the reducing concentrations across all scenarios.

Here we have compared the representations of total $PM_{2.5}$, primary $PM_{2.5}$ and secondary inorganic aerosol (SIA) concentrations, and how concentrations may be expected to reduce by 2040. These components

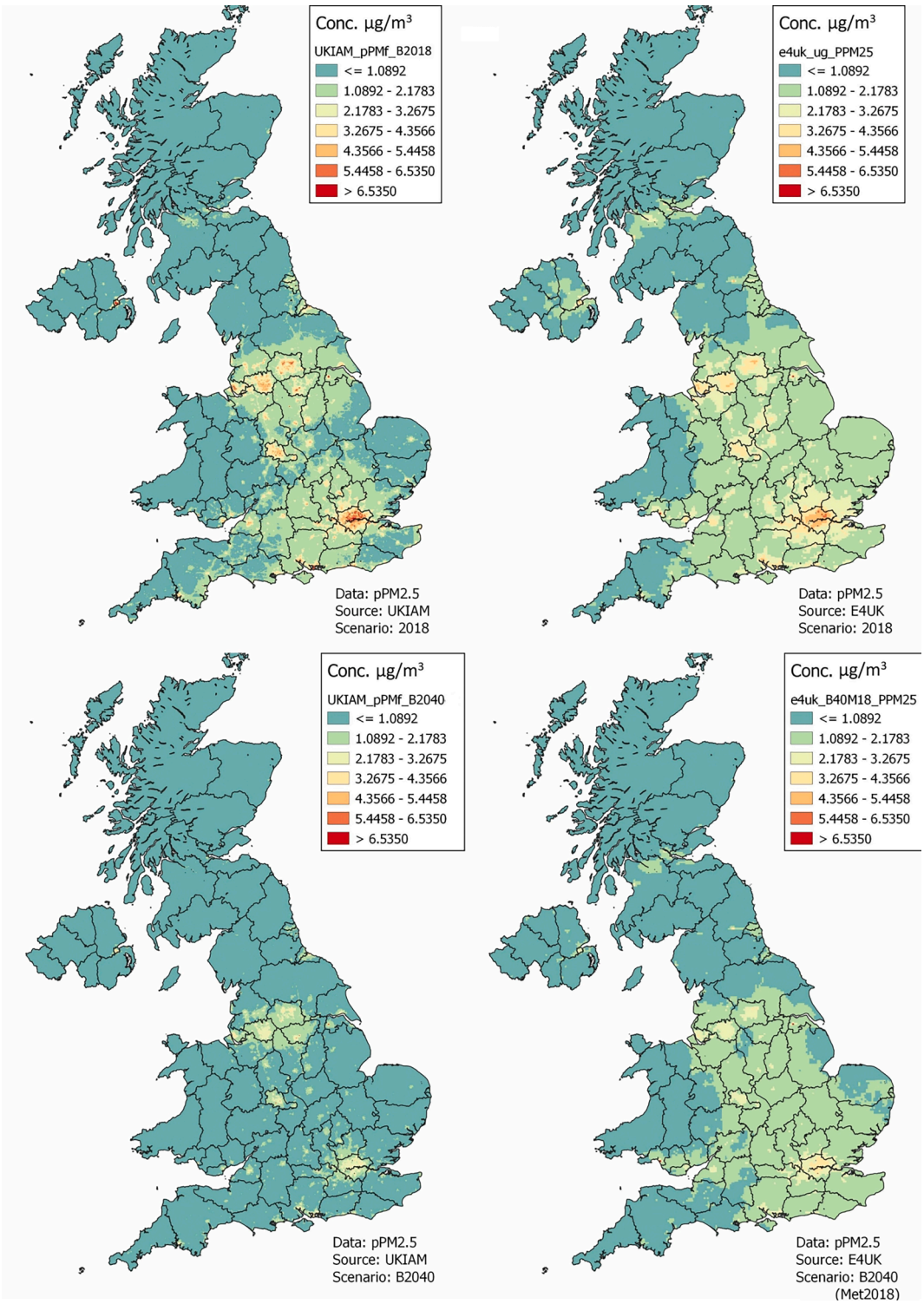

Fig. 5. Comparison of UKIAM and EMEP4UK for primary $PM_{2.5}$ concentrations in B2018 (upper) and for 2040 baseline conditions (lower).

Table 3
Comparison of Population Weighted Mean Concentration values for primary $PM_{2.5}$ in 2018.

PWMC µg m^{-3}	National	London	Urban	Rural
UKIAM	2.8	4.9	3.2	1.5
EMEP4UK	2.7	4.6	2.9	1.9

can be reduced through reductions in anthropogenic emissions. However, total $PM_{2.5}$ also includes natural contributions which are both uncertain and difficult to model. One such component is Secondary Organic Aerosol (SOA), which remains static in the UKIAM modelling and shows little change in the EMEP4UK modelling. In relation to natural dusts the models also differ, but the contributions assumed in each model are consistently in the region of 2 µg m^{-3} population weighted mean concentration (see Fig. S.6).

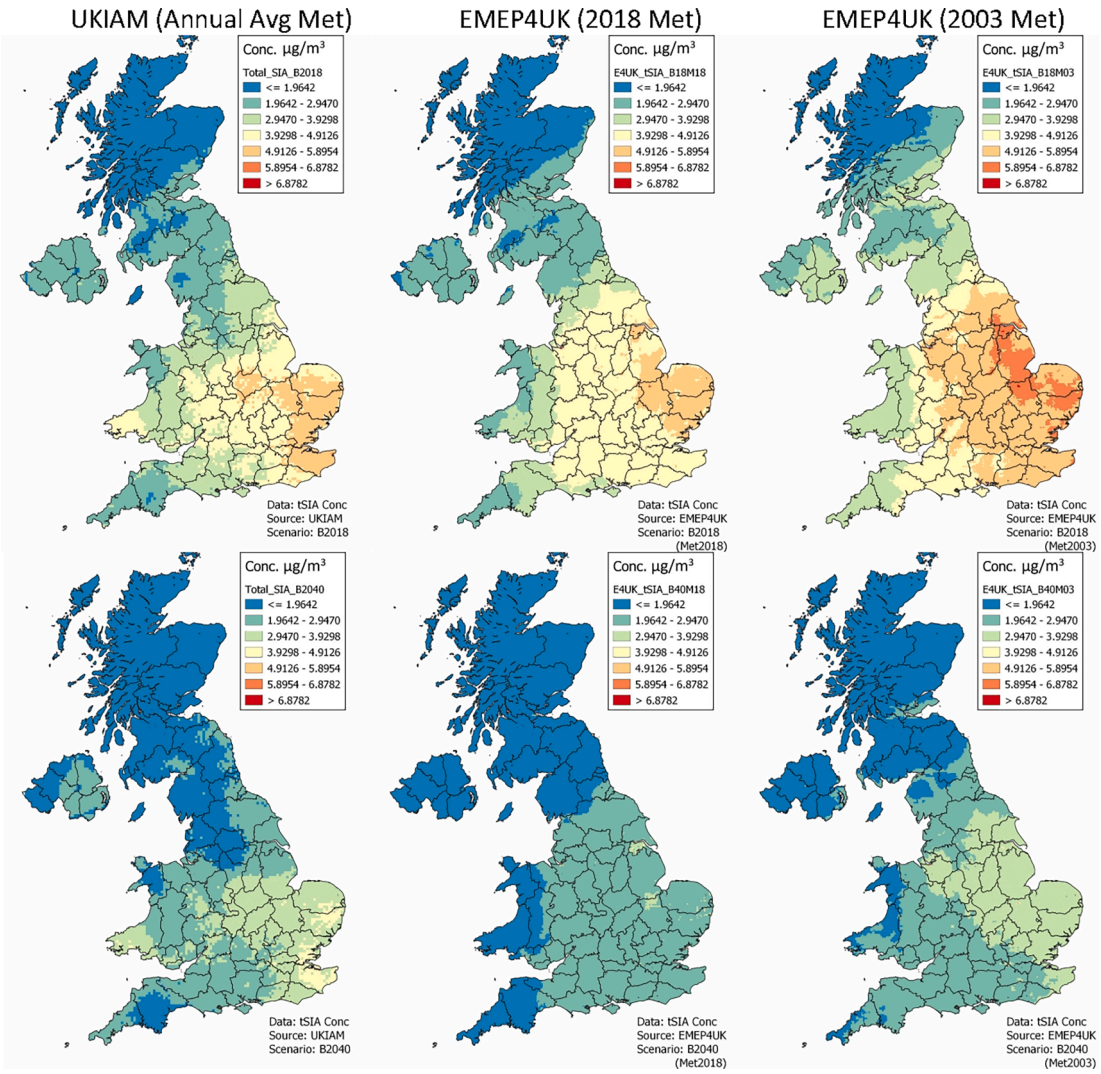

Fig. 6. Comparison of SIA concentrations in 2018 (upper) and for 2040 baseline conditions (lower). EMEP4UK concentrations are shown for different meteorological years (2018 and 2003).

4.3. Uncertainties and limitations

Both models contain uncertainties which may relate to input data, representation of processes, assumptions about natural contributions, variable meteorology, sources missing from emission inventories or spatial resolution. For example, the use of source-receptor relationships in UKIAM affects the ability to capture non-linearities in SIA chemistry, whereas reduction of emissions in EMEP4UK are calculated at the SNAP level (as opposed to sub-SNAP level), which may suppress the magnitude of emission changes at the local level.

These uncertainties have been discussed in detail elsewhere (ApSimon et al., 2021a; ApSimon et al., 2022). It should be recognised that whereas both models compare well with measurements, some uncertainties in the models may have a compensatory influence. For example, whereas emissions from cooking are not included in the NAEI and may contribute between 1 and 2 µg m^{-3} across London (see Oxley et al., 2020), it is also understood that estimates of domestic combustion of wood may have been overestimated (Oxley & ApSimon, 2018). A strength of the UKIAM is that sensitivity studies can be rapidly evaluated to understand such uncertainties, adding to the confidence in interpreting results of modelling projected futures; extensive sensitivity studies of this nature are reported by ApSimon et al., (2022).

It has been noted that the two models represent SIA concentrations in a different way. Whereas the UKIAM assumes linear scaling of source-receptor relationships in relation to emission changes, combined with a static adjustment based upon measurements from the AGANET monitoring network, the EMEP4UK model calculates SIA explicitly via the thermodynamic equilibrium with gas-phase precursors and will be affected by meteorology. In relation to SOA concentrations, UKIAM assumes a static representation calculated by the NAME model (Redington & Derwent, 2013) whereas EMEP4UK models SOA directly, albeit with uncertainties relating to IVOC/SVOC's not included in the inventory.

Other uncertainties to note, but which are not addressed in detail here, are uncertainties in non-exhaust emissions from road transport, especially in relation to progressive electrification of the fleet (Mehlig et al., 2021), or natural contributions from sea salt, Sahara dust, and other windblown dusts (see Fig. S.5). Overall however, all these uncertainties can be addressed by one model or the other through sensitivity studies (for example, for woodburning (Oxley & ApSimon, 2018)), and will benefit interpretation of results in relation to future policy scenarios.

Both models have limitations, and the comparisons presented here are based on national scale modelling and should therefore not be interpreted as being applicable to limited locations or timescales which may be at finer spatial or temporal resolutions than captured by either model.

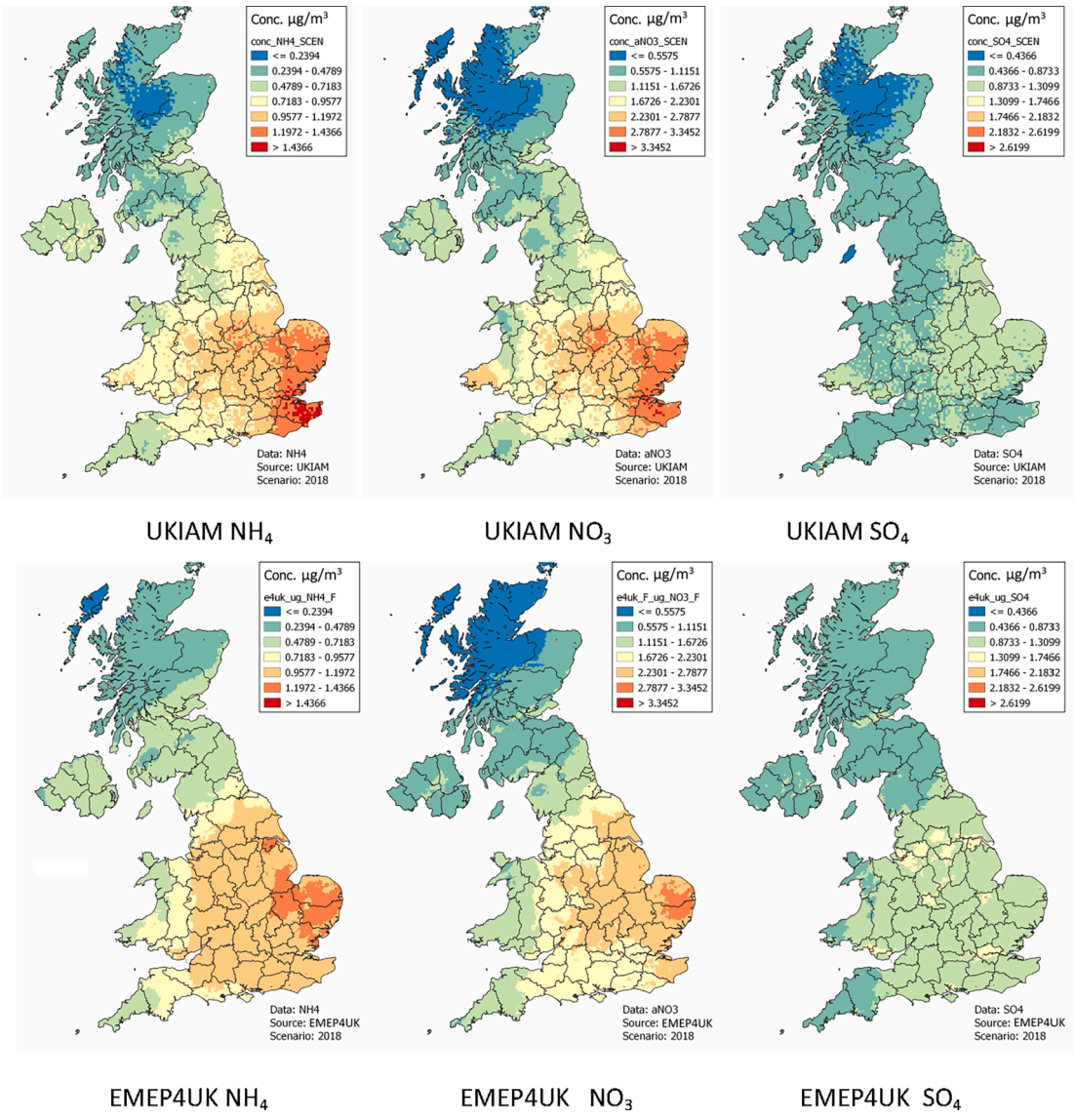

Fig. 7. Comparison of SIA components for 2018.

Table 4
PWMC values for individual SIA components (2018 & 2040), with EMEP4UK using 2018 meteorology.

	PWMC ($\mu g m^{-3}$)	National	London	Urban	Rural
B2018	UKIAM NO_3^-	2.07	2.75	2.1	1.97
	UKIAM NH_4^+	0.93	1.2	0.94	0.89
	UKIAM SO_4^{2-}	0.81	0.86	0.82	0.79
	E4UK NO_3^-	1.91	2.03	1.93	1.85
	E4UK NH_4^+	1	1.09	1.01	0.94
	E4UK SO_4^{2-}	1.18	1.35	1.21	1.07
B2040	UKIAM NO_3^-	1.44	1.91	1.45	1.41
	UKIAM NH_4^+	0.72	0.95	0.73	0.69
	UKIAM SO_4^{2-}	0.52	0.57	0.52	0.51
	E4UK NO_3^-	1.11	1.16	1.12	1.07
	E4UK NH_4^+	0.57	0.61	0.58	0.54
	E4UK SO_4^{2-}	0.66	0.74	0.67	0.61

5. Conclusions

The purpose of this work has been twofold. Firstly, to show how a combination of both reduced-form and complex ACTM's can be used in support of air quality policy development (see Fig. 1), and secondly, confirming the robustness of the UK Integrated Assessment Model for assessing alternative futures through a direct comparison with the Eulerian EMEP4UK model. Both models have shown good agreement with measurements (Figs. 2 & 3), EMEP4UK shows an ability to replicate past trends (Fig. S.1), and both models have been reviewed by the Air Quality Expert Group (AQEG, 2021).

The contrasting strengths of each model ensure that evaluation of the impacts of different projected futures will be an iterative process, whereby emerging scientific evidence can enhance understanding and result in increased confidence in model results and the development of robust policy scenarios.

With these considerations kept in mind, and noting the uncertainties and limitations discussed above (Section 4.3), the results of the comparisons between the UKIAM and EMEP4UK presented here show good agreement, with a clear understanding of why the models may show differences. The comparisons have included total $PM_{2.5}$, primary $PM_{2.5}$ and Secondary Inorganic Aerosol (SIA); the remaining 'other' contributions show a variation of approximately $\pm$ 0.2 µg m^{-3} PWMC between models (see Fig. S.5) and are the subject of ongoing investigation.

Whereas both models agree well (assuming 2018 meteorology in EMEP4UK) in 2018, showing a PWMC of 9.2 µg m^{-3}, it is clear that the UKIAM appears more conservative in relation to reductions of total

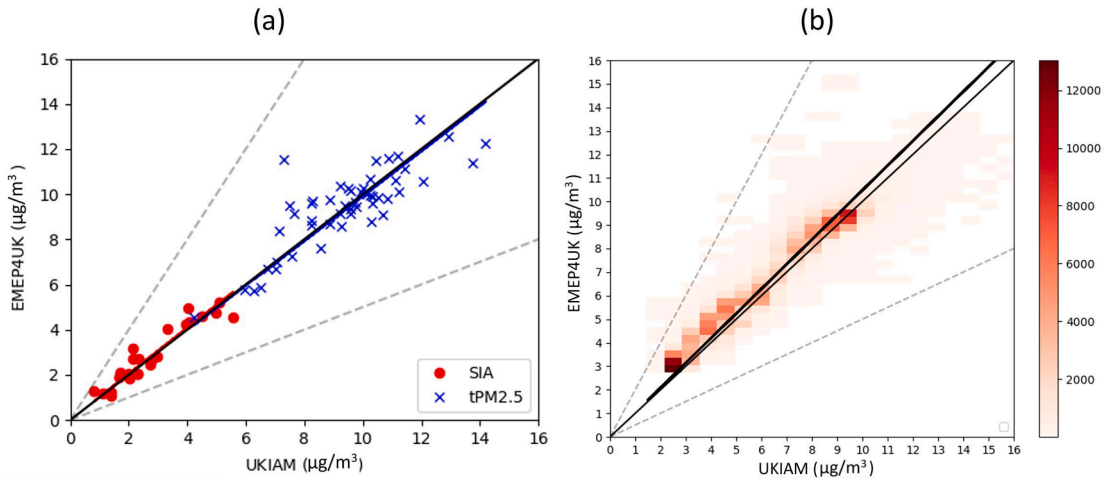

Fig. 8. A comparison of model results for 2018 for (a) SIA and total $PM_{2.5}$ concentrations at monitoring locations, and (b) a cell-by-cell (1 × 1 km^2) comparison of total $PM_{2.5}$ concentrations across the full model domain. The heat plot quantifies the number of cells (from a total of 244,387) showing the specified concentration. See Supplementary Information for comparison statistics and 2040.

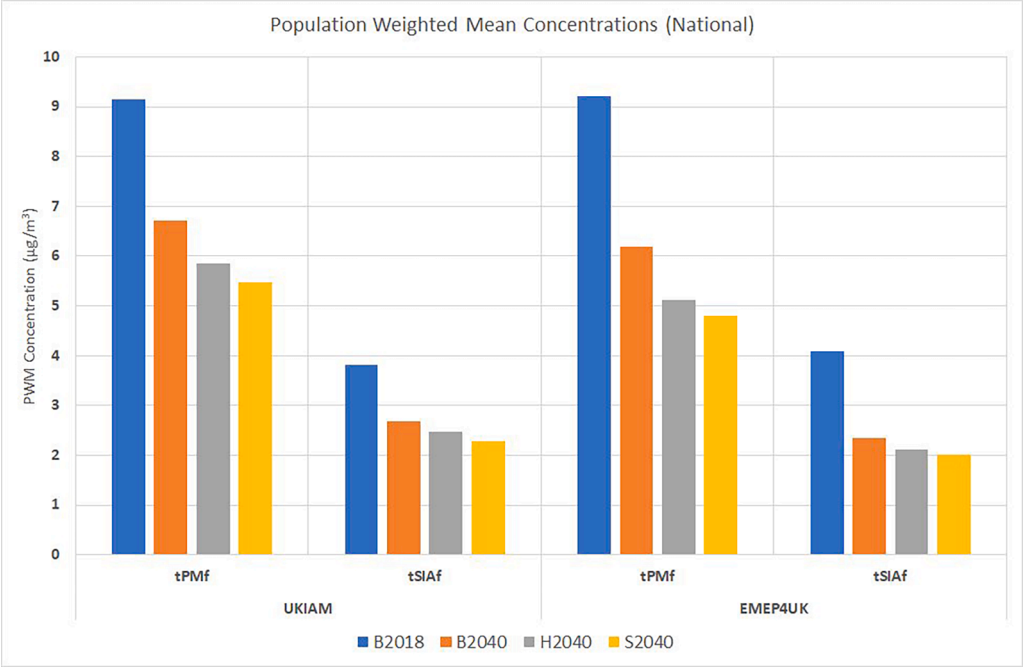

Fig. 9. A comparison of modelled total $PM_{2.5}$ and SIA population weighted mean concentrations for 2018 and 2040 (Baseline, High ambition, & Speculative) scenarios, showing good agreement between the models.

$PM_{2.5}$ concentrations by 2040. Whereas UKIAM shows a reduction to 6.7 µg m^{-3}, EMEP4UK shows a reduction to 6.2 µg m^{-3}. Similarly, where UKIAM shows a reduction in SIA concentrations of 0.9 µg m^{-3}, EMEP4UK reduces SIA by 1.8 µg m^{-3}.

The other contribution to total $PM_{2.5}$ concentrations reported above is the *primary* $PM_{2.5}$. As in all the scenarios modelled, the total UK emissions are consistent between the models. However, EMEP4UK is more conservative in reductions of concentrations by 2040 (see Fig. 5). The reasons for this are twofold. Firstly, the UKIAM has a higher resolution of 1 × 1 km^2 which will result in larger concentration peaks in urban areas, and emission abatement measures can be applied to specific sources. Secondly, emissions for EMEP4UK are scaled at the level of SNAP sectors, as opposed to the sub-SNAP level sources captured by the UKIAM. This means that reductions in emissions may effectively be spread over a number of different sources.

Overall, and considering the uncertainties involved, the comparisons presented above suggest good agreement of concentrations of $PM_{2.5}$ for both the current situation and projected future scenarios, giving confidence in the results generated by both models. The models are always evolving to capture the latest scientific understanding, and future developments should address some of the uncertainties discussed above. For example, although the static SIA adjustment assumed by the UKIAM has been acknowledged as being beneficial (AQEG, 2021), further work is recommended to investigate how future changes to atmospheric composition may affect this parameterisation.

Finally, these comparisons have highlighted how a combination of reduced-form modelling (UKIAM) and complex chemical transport modelling (EMEP4UK) can be effectively used in support of air pollution policy development (see ApSimon et al., 2022), informing understanding of projected futures in the context of emerging evidence and

uncertainties (see Fig. 1).

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CRediT authorship contribution statement

Tim Oxley: Conceptualization, Methodology, Software, Validation. **Massimo Vieno:** . **Huw Woodward:** Methodology, Data curation. **Helen ApSimon:** Conceptualization, Methodology, Project administration. **Daniel Mehlig:** . **Rachel Beck:** Data curation. **Eiko Nemitz:** . **Stefan Reis:** .

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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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 A. Supplementary material

Supplementary data to this article can be found online at https://doi.org/10.1016/j.envint.2022.107676.

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Glossary

ACTM: Atmospheric Chemistry Transport Model (eg. EMEP4UK)
AGANET: Acid Gas and Aerosol Network of monitoring stations, https://uk-air.defra.gov.uk/networks/network-info?view=aganet
AQEG: Air Quality Expert Group, https://uk-air.defra.gov.uk/research/aqeg/
ASAM: Abatement Strategies Assessment Model capturing transboundary contributions to UK air pollution (ApSimon et al., 1994)
AURN: Automatic Urban and Rural Network of monitoring stations, https://uk-air.defra.gov.uk/networks/network-info?view=aurn
BRUTAL: Road Transport sub-model of the UKIAM (Oxley et al., 2009)
CLRTAP: UNECE Convention on Long-Range Transboundary Air Pollution; renamed as the Air Convention https://unece.org/environment-policy/air
CORINAIR: CORe INventory of AIR Emissions
EMEP: (1) Cooperative Programme for Monitoring and Evaluation of the Long-range Transmission of Air Pollutants in Europe (1984, Geneva Protocol) https://www.emep.int/ (2) Unified EMEP Eulerian model (Simpson et al., 2012; https://github.com/metno/emep-ctm
EMEP4UK: EMEP4UK model (Vieno et al., 2009; 2010; 2014; 2016)
GAINS: Greenhouse gas and Air pollution INteractions and Synergies; a development of the RAINS model to address the inter-relationships with effects of greenhouse gases (GHG), https://gains.iiasa.ac.at/models/
MARGA: Monitor for AeRosols and Gases in Air (https://www.pollutantdeposition.ceh.ac.uk/emep)
NAEI: National Atmospheric Emissions Inventory (https://naei.beis.gov.uk)
NAMN: National Ammonia Monitoring Network (https://www.pollutantdeposition.ceh.ac.uk/aganet)
NH_3: Ammonia
NH_4^+: Ammonium Aerosol, forming either ammonium nitrate (NO_3NH_4) or ammonium sulphate ($SO_4(NH_4)_2$)
NO_3^-: Nitrate Aerosol (in this paper this always refers to the fine (<2.5μm) NO_3^-)
NOx: Nitrogen Oxides comprised mainly of NO (Nitric Oxide) and NO_2 (Nitrogen Dioxide)
$PM_{2.5}$: Particulate Matter < 2.5μm diameter
PWMC: Population Weighted Mean Concentration, $PWMC = \sum_{ij}(P_{ij} \times C_{ij})/\sum_{ij}P_{ij}$, where the population in cell (ij) is P_{ij} and the concentration is C_{ij}
SIA: Secondary Inorganic Aerosol, formed by precursor emissions of NH_3, SO_2 and NOx ($SIA=SO_4^{2-}+NO_3^-+NH_4^+$)
SNAP: Selected Nomenclature for Air Pollutants
SOA: Secondary Organic Aerosol, influenced by both biogenic and anthropogenic emissions
SO_2: Sulphur Dioxide
SO_4^{2-}: Sulphate Aerosol
UKIAM: UK Integrated Assessment Model, developed by Imperial College London (ApSimon et al., 2021a; Oxley et al., 2013)
VOC: Volatile Organic Compounds