

2.13 REGIONAL SCALE MODELLING OF PARTICULATE MATTER IN THE UK: ATTRIBUTION AND FUTURE TRENDS

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INTRODUCTION

Particulate matter is of concern because of its established relationship with health effects on the cardiovascular and respiratory systems. The focus in health terms is on fine particles (PM₁₀ and smaller), which are able to enter the respiratory tract. These effects apparently relate to both short term and long term exposure (COMEAP 1998 and 2001). For urban areas in the UK, it has been suggested that exposure to PM₁₀ can result in an additional 10,500 additional (or early) hospital admissions and 8,100 deaths brought forward. Air quality standards for PM₁₀ have been set by global (the WHO), regional (the EU) and national (UK) institutions. In the latest amendments to the UK's Air Quality Strategy (2003) different regional targets have been set, with the standard for the annual mean ranging from 18 µg m⁻³ for Scotland to 23 µg m⁻³ for London (to be achieved by 2010). The standard for the rest of England, Wales and Northern Ireland is 20 µg m⁻³.

Particles are made up of a wide range of materials, of varying size and composition. There are three main source types: primary (from combustion, including road traffic); secondary (products of oxidation in the atmosphere) and coarse particles of both natural (sea salt, dust) and anthropogenic (tyre wear, quarrying) origin. As both the size and composition of particles seems to determine their health effects, it is important to be able to identify the different components (sources) of measured particulates as this will help to target emissions reduction policies. In the UK, PM₁₀ are mapped for background (with a constant coarse particle concentration of 8.8 µg m⁻³ (gravimetric)) and roadside. Source apportionment is based on the APEG model (APEG, 1999; Stedman *et al.*, 2003), using regression analysis.

In this paper we present a different approach to estimating annual average particulate concentrations across the UK by using two regional scale models, HARM (Hull Acid Rain Model) and ELMO (Edinburgh Lancaster Model for Ozone). The models are used to generate estimates for 1999 including primary PM₁₀ and secondary inorganic and organic components. A regional coarse component is added to facilitate direct comparison with the outputs of the NETCen GIS-based model (Stedman *et al.*, 2003) and the Unified EMEP model (Kahnert and Tarrasón, 2003). HARM and ELMO are then used to simulate particulate concentrations across the UK in 2010 when EU and UNECE member states have reduced emissions of SO₂, NO_x, NH₃ and VOCs in line with their commitments to the EU National Emissions Ceilings Directive and the Gothenberg Protocol.

THE MODELS

HARM and ELMO are both receptor-orientated Lagrangian trajectory models. The model domain is the same as the EMEP model, using EMEP emissions data outside the area covered by the UK OS grid. Inside this area, the models employ emissions data from the National Atmospheric Emissions Inventory (NAEI), aggregated to 10 km x 10 km resolution. The structure and chemistry of both models have been described in detail elsewhere (Metcalfe *et*

al., 2001, 2002). Here we are using a new version of HARM (HARM12.1) which includes global background concentrations, multiple vertical layers with vertical diffusion between them and ecosystem-specific deposition (Metcalf *et al.*, in prep.). Primary PM₁₀ inventories for EMEP and the UK have been included. The particles are assumed to be chemically inert and are simply emitted, mixed, transported and removed. HARM is used to estimate the concentrations of primary PM₁₀ and secondary inorganic aerosols resulting from emissions of SO₂, NO_x and NH₃. ELMO is used to model secondary organic aerosol through the implementation of a condensed version of the α -pinene photo-oxidation scheme taken from the STOCHEM model. It adopts 50 km x 50 km estimates of isoprene and monoterpene emissions across the EMEP grid domain and 10 km x 10 km estimates of isoprene and monoterpene emissions across the GB based on inventories recently developed by Stewart *et al* (2003). Both models have been run using 1999 emissions data and precipitation data. For the 2010 model runs, in addition to national and EU data, we have scaled UK and EMEP 1999 PM₁₀ emissions to 2010 levels based on country totals published by Klimont *et al* (2002).

MODEL VALIDATION

Modelled primary PM₁₀ concentrations from HARM have been compared with data from the UK's Smoke and SO₂ Network using simple linear regression. Sample output is illustrated in Figure 1. The model does not appear to reproduce either the amount of particulate mass or its variation across the country very effectively ($R^2 = 0.176$). However, there are issues concerning the precision of present day black smoke measurements, hence the modelled output has been adopted with caution for the purposes of this study.

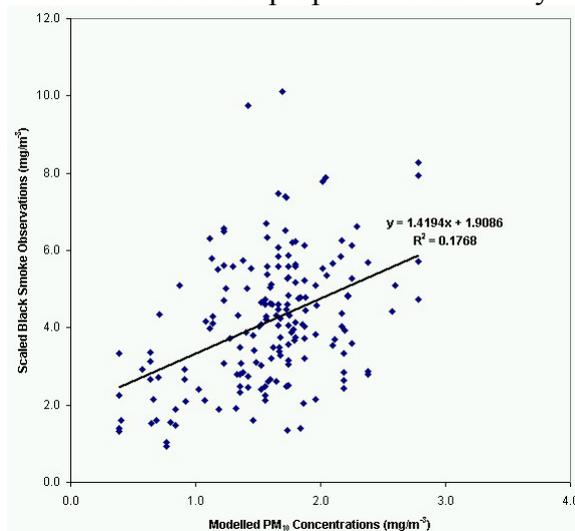


Figure 1. Regression of HARM modelled primary PM₁₀ concentrations (in $\mu\text{g m}^{-3}$) against data from the UK's Smoke and SO₂ monitoring network

Modelled secondary inorganic aerosol concentrations from HARM have been compared with data from the UK's National Nitric Acid Monitoring Network for SO₄²⁻, NO₃⁻, HNO₃ and NH₄⁺ using linear regression. Sample output is illustrated in Figure 2. The analysis shows that for SO₄²⁻, NO₃⁻ and NH₄⁺ HARM is able to reproduce both the amount and spatial distribution of aerosol well (although with some underestimation of high concentrations of NH₄⁺). The model also reproduces the spatial pattern of nitric acid, but underestimates substantially. Based on these results, it seems reasonable to use HARM to model the secondary inorganic component of PM₁₀.

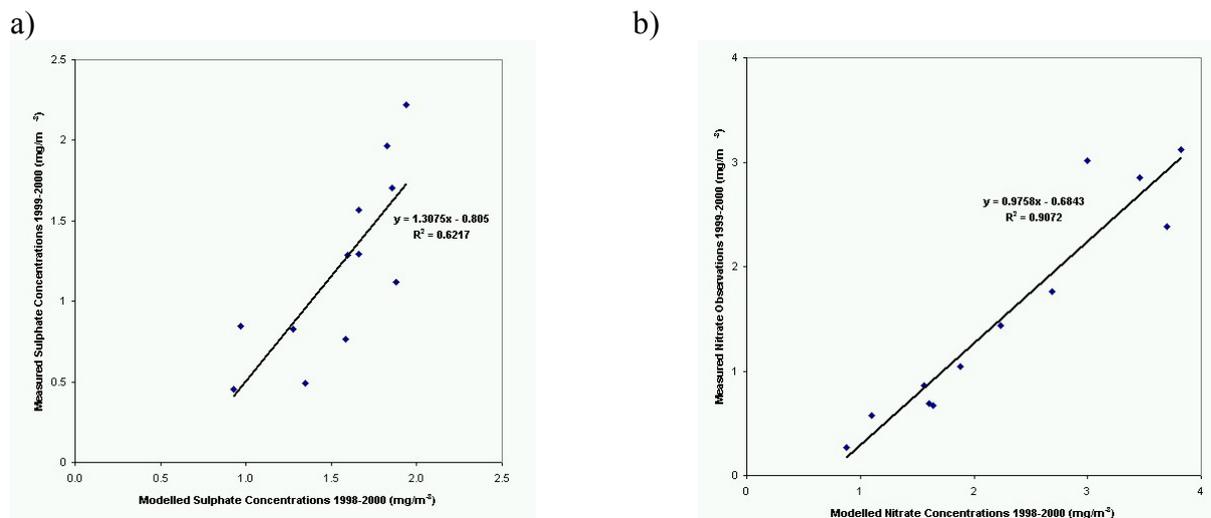


Figure 2. Regression of the HARM modelled aerosol concentrations (in $\mu\text{g m}^{-3}$) against data from the UK's National Nitric Acid Monitoring Network a) Sulphate Aerosol b) Nitrate Aerosol

Secondary organic aerosol concentrations are not routinely monitored across the UK hence a data model comparison has not been possible.

Primary inorganic and secondary organic and inorganic components derived from HARM and ELMO have been combined with a coarse regional component of $8.8 \mu\text{g m}^{-3}$ gravimetric to produce an estimate of annual average particulate concentrations across the UK. This is directly comparable with estimates from the NETCen GIS-based model (Figure 3).

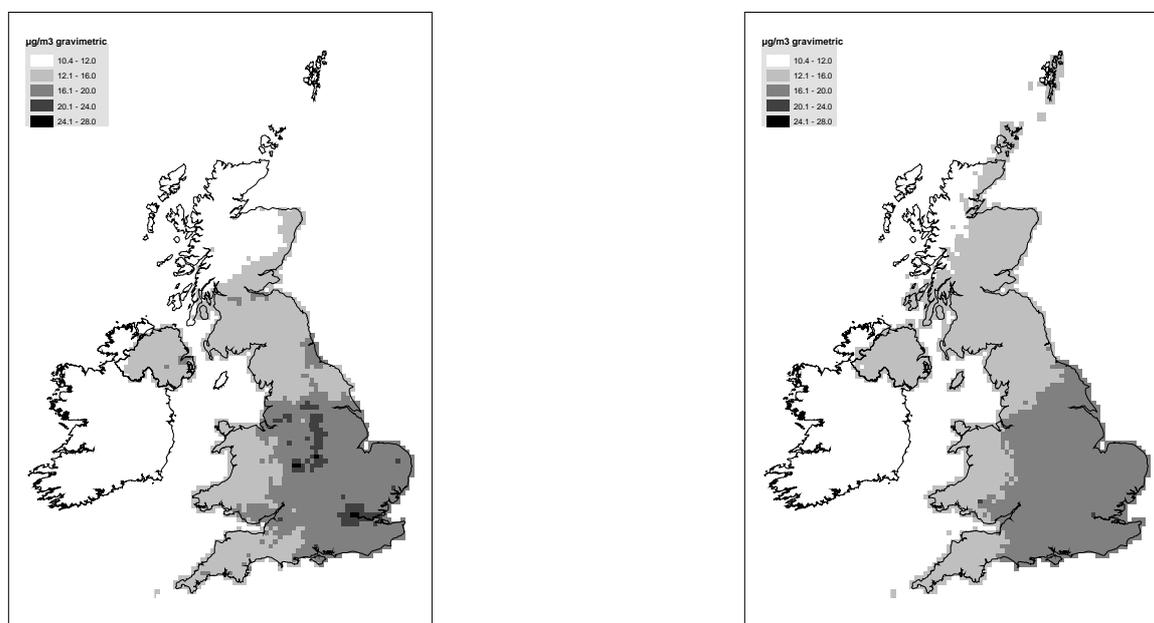


Figure 3. Modelled Average Annual Particulate Concentrations (in $\mu\text{g m}^{-3}$) derived from a) the NETCen model (2002) and b) Combined HARM and ELMO outputs (1999)

The HARM and ELMO estimate of concentrations compares favourably against the NETCen estimate of background concentrations in remote regions but tends to under-predict close to major urban areas. Given that the model is able to reproduce levels of secondary inorganic aerosol quite effectively (Figure 2) this under-prediction close to major urban areas must be associated with the primary component of the particulate load. Comparison with measurements from the urban PM₁₀ network confirms that this is indeed the case. One possible explanation for this under-estimation is model scale. HARM operates at a spatial scale of 10 km which is too coarse to effectively reproduce particulate concentrations in urban area. A detailed comparison with the output of the Unified EMEP model has not been possible at this stage, however, the model results presented here appear to be of similar magnitude but higher spatial variability than those produced by the Unified EMEP model (Kahnert and Tarrasón, 2003)

SOURCE ATTRIBUTION AND THE EFFECTS OF EMISSIONS REDUCTIONS

The modelled attribution of particulates at the UK National Nitric Acid Monitoring Network sites is depicted in Figure 5. This attribution excludes the constant coarse component ($8.8 \mu\text{g m}^{-3}$) in order to highlight regional variations in source attribution.

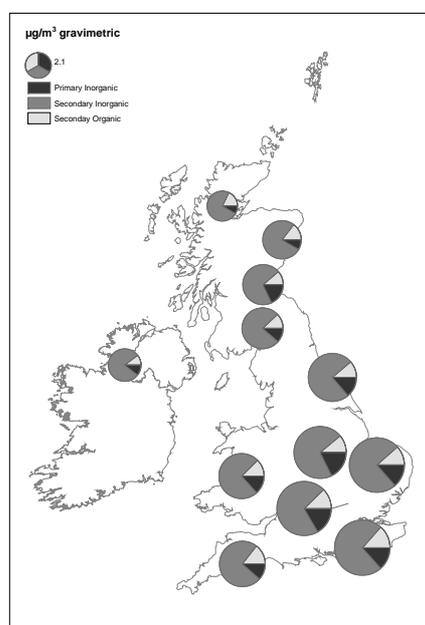


Figure 5. Modelled concentrations and source attribution of PM₁₀ in the UK (1999) based on HARM and ELMO output

The modelling indicates that secondary inorganic aerosol is the main component of PM₁₀ at all sites (70 - 84%). There are four sites (Glensaugh and Strathvaich Dam in Scotland, Eskdalemuir in northern England, and Yarnar Wood in south west England) where the contribution from secondary organic aerosol is larger than that from primary PM₁₀. As these are all sites remote from major source areas, this is unsurprising. At Lough Navar in Northern Ireland, the two contributions are about equal.

Running the models using 2010 emissions (NECD) only has a small impact on total PM₁₀ concentrations. This is perhaps, not too surprising, as 1999 emissions are quite close to their 2010 targets in many cases. Although secondary inorganic aerosol continues to be the dominant source category, the relative importance of secondary organic aerosols increases,

becoming the second most important source category at all sites except Bush (central valley of Scotland) and Sutton Bonnington (East Midlands of England).

CONCLUSIONS

Output from HARM and ELMO has been used to derive estimates of total PM₁₀ across the UK. The modelled output shows a good level of agreement with the NETCen estimates in rural areas where concentrations are dominated by the secondary inorganic component, but poor levels of agreement in urban areas where the primary component dominates. Further model development is clearly necessary if HARM is to provide an effective assessment of the primary fraction.

On the basis of the output presented here few areas of the country exceeded annual mean standards for PM₁₀ in 1999. This is not surprising given the under-estimation of the primary component. By 2010 all parts of the UK fall comfortably below 20 µg m⁻³, indicating that substantial reductions in primary and secondary inorganic aerosol may be achieved through emission reduction across Europe. However, these results also need to be interpreted with caution given the known under-estimation of the primary component.

In this study secondary organic aerosol has been included as a component of total PM₁₀. At present, there is little monitoring data with which to compare modelled estimates, however, on the basis of results presented here, the secondary organic component contributes between 10-15% of the total particulates load in 1999 and 13-20% in 2010 if the coarse regional component is discounted. Clearly the relative importance of this component will only increase as emissions from anthropogenic sources decrease in response to national and international policy.

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