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**LETTER**

## The UK particulate matter air pollution episode of March–April 2014: more than Saharan dust

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## Abstract

A period of elevated surface concentrations of airborne particulate matter (PM) in the UK in spring 2014 was widely associated in the UK media with a Saharan dust plume. This might have led to over-emphasis on a natural phenomenon and consequently to a missed opportunity to inform the public and provide robust evidence for policy-makers about the observed characteristics and causes of this pollution event. In this work, the EMEP4UK regional atmospheric chemistry transport model (ACTM) was used in conjunction with speciated PM measurements to investigate the sources and long-range transport (including vertical) processes contributing to the chemical components of the elevated surface PM. It is shown that the elevated PM during this period was mainly driven by ammonium nitrate, much of which was derived from emissions outside the UK. In the early part of the episode, Saharan dust remained aloft above the UK; we show that a significant contribution of Saharan dust at surface level was restricted only to the latter part of the elevated PM period and to a relatively small geographic area in the southern part of the UK. The analyses presented in this paper illustrate the capability of advanced ACTMs, corroborated with chemically-speciated measurements, to identify the underlying causes of complex PM air pollution episodes. Specifically, the analyses highlight the substantial contribution of secondary inorganic ammonium nitrate PM, with agricultural ammonia emissions in continental Europe presenting a major driver. The findings suggest that more emphasis on reducing emissions in Europe would have marked benefits in reducing episodic $PM_{2.5}$ concentrations in the UK.

## 1. Introduction

An air pollution event of elevated surface concentrations of particulate matter (PM) occurred in the UK between 26 March and 8 April, 2014. Observations from UK air quality monitoring networks showed markedly elevated PM surface concentrations across the majority of the country during this time. The UK Automatic Urban and Rural monitoring Network (AURN) recorded hourly values of $PM_{10}$ of up to 100 $\mu g\,m^{-3}$, well above the hourly concentrations of a few tens $\mu g\,m^{-3}$ normally recorded; for example across 45 urban background AURN sites the median, inter-quartile range and 95th percentile of hourly

$PM_{10}$ concentrations in 2014 were, respectively, 38, 36–44 and 53 $\mu g\,m^{-3}$ (http://uk-air.defra.gov.uk).

The spring 2014 PM episode was widely perceived as being associated with an enhanced surface concentration of mineral dust from the Sahara (e.g. as reported by the BBC[6], the Guardian[7], and the Daily Mail[8]). An implication is that this was, at least in part, a 'natural' event. However, it is known that the UK's

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[6] www.bbc.co.uk/news/uk-26844425.
[7] www.theguardian.com/uk-news/2014/apr/01/london-smog-saharan-dust-storms-downing-street.
[8] www.dailymail.co.uk/wires/ap/article-2594987/Sahara-desert-dust-brings-smog-Britain.html.

geographic proximity to industrialised countries in Western Europe (e.g. Germany, France and the Netherlands) make it vulnerable to the import of anthropogenic primary and secondary gaseous pollutants and PM . Previous analyses have highlighted this issue for ozone (Vieno *et al* 2010, Francis *et al* 2011) and for secondary inorganic components of PM (Abdalmogith and Harrison 2005, Jones and Harrison 2006, Yin and Harrison 2008, Charron *et al* 2013, Vieno *et al* 2014). For effective policy action to mitigate the impacts of PM on human health it is essential to understand the composition of the atmosphere and the causes of high levels of PM (e.g. Heal *et al* 2012), of which this episode was an example. Of the studies investigating the origins of elevated PM cited above, all except Vieno *et al* (2014) were based on analysis of measurements of PM chemical components at Harwell or Birmingham in south-central UK. The key feature of the current work was the combination of speciated PM measurements, which are only sparsely undertaken in the UK (as elsewhere), and a regional 3D Eulerian atmospheric chemistry transport model (ACTM) that simulates to 5 km horizontal resolution over the region of study, to assess the validity of attributing this PM event to Saharan dust over the entire UK domain.

## 2. Method

### 2.1. Atmospheric chemistry transport modelling

A Eulerian ACTM, the EMEP4UK version rv4.3 (Vieno *et al* 2010, Simpson *et al* 2012, Vieno *et al* 2014, Vieno *et al* 2016), was used to study the spring 2014 UK air pollution event. The meteorological driver used for EMEP4UK for this purpose was the Weather Research and Forecasting (WRF) model version 3.6. The WRF model included data assimilation (Newtonian nudging) of the numerical weather prediction model meteorological reanalysis from the US National Center for Environmental Prediction/National Center for Atmospheric Research Global Forecast System at 1° resolution, every 6 hours (Saha *et al* 2011). The model domain covers the whole of Europe at a horizontal resolution of 50 km × 50 km with a higher horizontal resolution nested inner domain over the British Isles at 5 km × 5 km, as shown in figure 1.
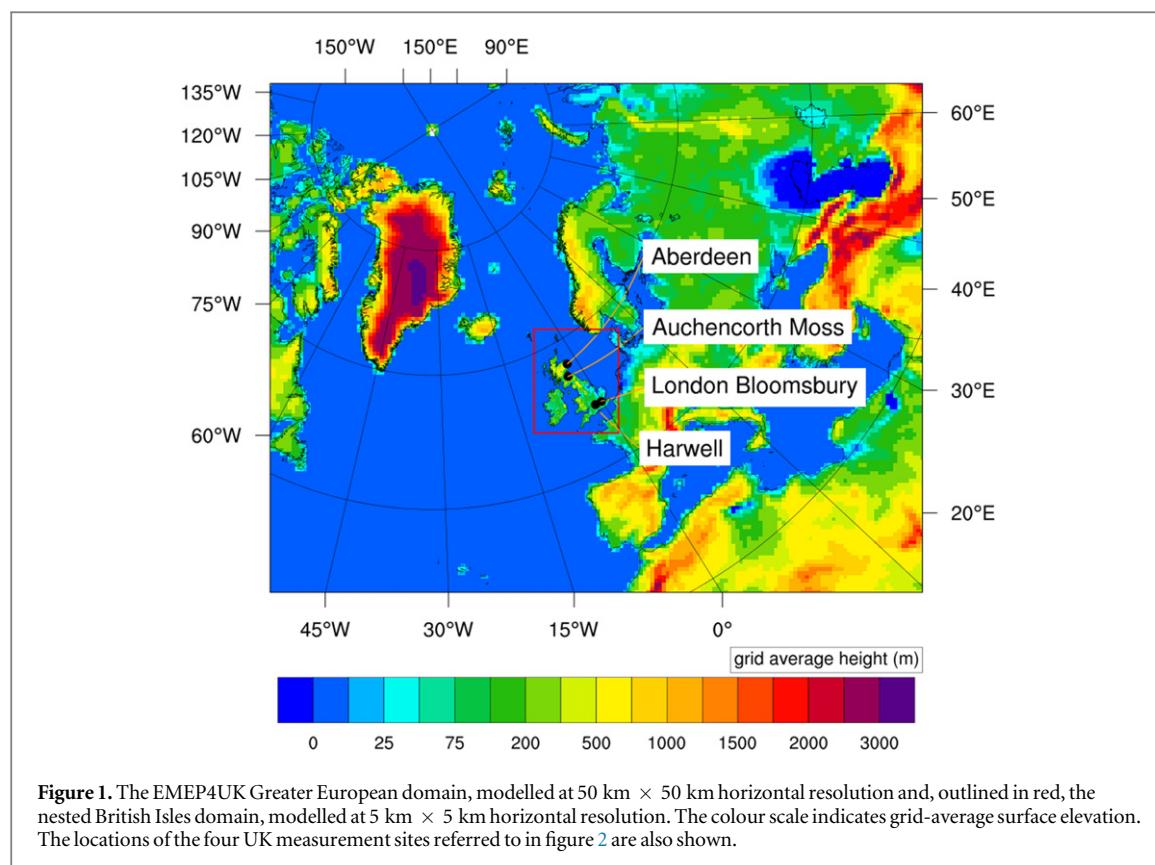
A full description of the EMEP4UK model can be found in Vieno *et al* (2014) and Simpson *et al* (2012). In addition to gaseous components, the EMEP4UK model simulates the concentrations (at hourly time resolution) of both $PM_{10}$ (particles with aerodynamic diameter $<10\,\mu m$) and $PM_{2.5}$ (particles with aerodynamic diameter $<2.5\,\mu m$), the two PM size fractions forming part of EU air quality legislation (Heal *et al* 2012). The chemical scheme used for the present study is the EMEP-EmChem09 (Simpson *et al* 2012, www.emep.int) and the gas/aerosol partitioning was the model for aerosols reacting system (MARS)

(Binkowski and Shankar 1995). In the model version used here, $PM_{2.5}$ is derived as the sum of the fine fraction of: ammonium ($NH_4^+$), sulphate ($SO_4^{2-}$), nitrate ($NO_3^-$), elemental carbon (EC), organic matter (OM), sea salt (SS), mineral dust, and 27% of the coarse nitrate. $PM_{10}$ is derived as the sum of $PM_{2.5}$ plus the coarse fraction of EC, OM, $NO_3^-$, SS, and dust. In the current version of the EMEP4UK model, the split between $PM_{2.5}$ and $PM_{2.5-10}$ for $NO_3^-$ is somewhat uncertain as discussed in Aas *et al* (2012). Fine $NO_3^-$ production is modelled using the MARS thermodynamic model and the coarse $NO_3^-$ formation in the current model is a function of humidity and $HNO_3$ using a parametrization derived from Riemer *et al* (2003), as described in Simpson *et al* (2012), but it is not explicitly linked to the surface area of the existing coarse aerosol. Both nitrate generation mechanisms compete for the same $HNO_3$ therefore constraining the total amount of nitrate produced.

Anthropogenic emissions of $NO_x$, $NH_3$, $SO_2$, primary $PM_{2.5}$, primary coarse PM, CO and non-methane VOC for the UK are derived from the National Atmospheric Emission Inventory (NAEI, http://naei.defra.gov.uk) aggregated to 5 km × 5 km resolution. The EMEP 50 km × 50 km resolution emission estimates provided by the Centre for Emission Inventories and Projections (CEIP, www.ceip.at/) are used for the European domain. Shipping emissions estimates, for the inner domain, are derived from ENTEC (2010).

The boundary conditions at the edge of the European domain (figure 1) are prescribed concentrations adjusted for each simulated year as described in Simpson *et al* (2012). The EMEP4UK model version used here uses the default climatological Saharan dust boundary conditions, based on monthly average dust concentrations for the year 2000 from the global CTM2 at the University of Oslo. This treatment may lead to an inaccurate representation of the source strength of Saharan dust for individual events. The atmospheric flow and deposition processes transporting the dust and controlling its distribution over Europe and the UK during the simulated period are fully represented within the model. For ozone, a 3D field from climatological ozone-sonde data-sets, modified monthly against clean-air surface observations was used as initial and boundary condition for the European domain.

Two EMEP4UK model simulations were carried out to quantify the contributions of UK and regional emissions to the episode of elevated surface PM: (1) A base-case run, called 'BASE' hereafter, and (2) a no-UK anthropogenic emissions run, called 'NoUK' hereafter. The difference between the BASE and the NoUK simulations therefore represents a quantitative measure of the enhancement of the UK PM concentration that is caused by domestic emissions over the European background. The percentage fraction of PM generated by UK emissions ($f_{UK}$) was calculated as:

**Figure 1.** The EMEP4UK Greater European domain, modelled at 50 km × 50 km horizontal resolution and, outlined in red, the nested British Isles domain, modelled at 5 km × 5 km horizontal resolution. The colour scale indicates grid-average surface elevation. The locations of the four UK measurement sites referred to in figure 2 are also shown.

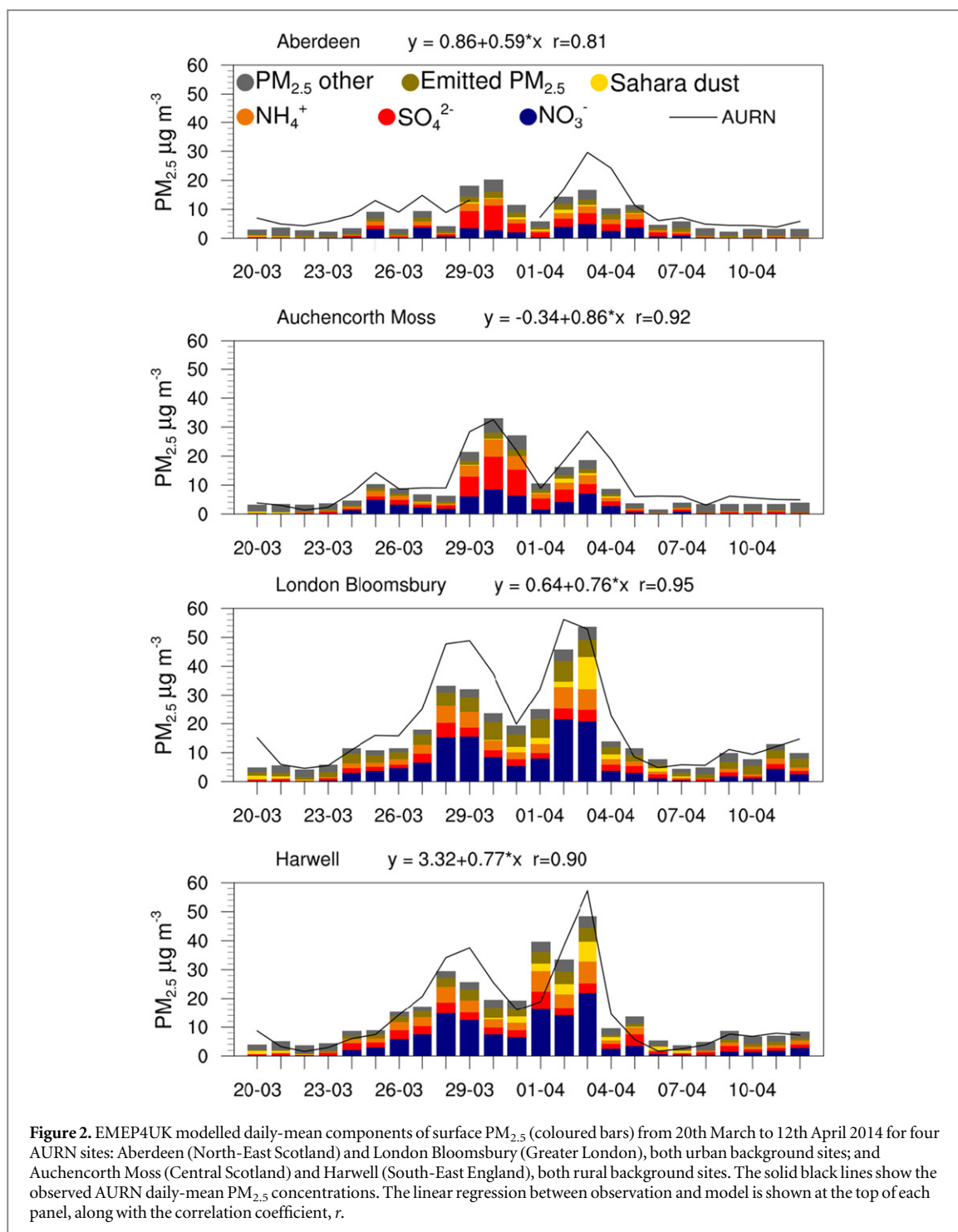$$f_{UK} = 100 \times (BASE - NoUK)/BASE. \quad (1)$$

## 2.2. Measurements

The daily-mean simulated surface concentrations of $PM_{10}$ and $PM_{2.5}$ across the UK were compared with data from the UK's AURN, which uses the Tapered Element Oscillating Microbalance Filter Dynamic Measurement System (TEOM-FDMS) (Bureau-Veritas 2010). Comparisons were undertaken for all AURN sites with $PM_{10}$ and $PM_{2.5}$ measurements; for the sake of simplicity this paper presents data for the following four representative sites (uk-air.defra.gov.uk): Aberdeen (North-East Scotland, lat 57.157°, long −2.094°, urban background); Auchencorth Moss (Central Scotland, lat 55.792°, long −3.243°, rural background); Harwell (South-East England, lat 51.571°, long −1.325°, rural background); and London Bloomsbury (Greater London, lat 51.522°, long −0.126°, urban background). The locations of these sites are marked on figure 1. The Harwell and Auchencorth Moss sites are part of the UKEAP (UK Eutrophying and Acidifying Atmospheric Pollutant) network. For this purpose, additional instrumentation includes a Monitor for AeRosols and GAses (MARGA, Metrohm Applikon B. V., NL), which measures hourly concentrations of the major secondary inorganic components of PM ($SO_4^{2-}$, $NO_3^-$, and $NH_4^+$), as described by Twigg *et al* (2015).

## 3. Results and discussion

The dominant meteorological feature for the duration of the PM pollution event was an area of high pressure located over Europe, with relatively low surface temperatures of 8 °C–14 °C. This limited the evaporation of ammonium nitrate into ammonia and nitric acid, enhancing the lifetime of particle nitrate and the potential influence of its long-range transport.

Figures 2 and 3 compare the modelled and measured daily mean surface concentrations of $PM_{2.5}$ and $PM_{10}$, respectively, between 20th March and 13th April 2014 at the four selected AURN sites. All four sites experienced elevated PM during this period when compared with their 2014 annual averages, which for $PM_{2.5}$ and $PM_{10}$ were: 9 and 14 $\mu g\,m^{-3}$ (Harwell), 15 and 19 $\mu g\,m^{-3}$ (London Bloomsbury), 10 and 15 $\mu g\,m^{-3}$ (Aberdeen) and 7 and 8 $\mu g\,m^{-3}$ (Auchencorth Moss) (uk-air.defra.gov.uk). The correlation coefficients between model and measurements over the simulation period are greater than 0.77 for all four sites included in this work for both $PM_{2.5}$ and $PM_{10}$ (figures 2 and 3).

Two distinct episodes are apparent in the $PM_{2.5}$ and $PM_{10}$ concentrations: one centred on the 30th March (referred to subsequently as episode 1: 'EP1') with an approximate duration of 5 days, and the second centred on the 3rd April ('EP2') with an approximate duration of 4 days. The spatial distributions across the UK and the rest of Western Europe of the daily mean surface PM during these episodes are
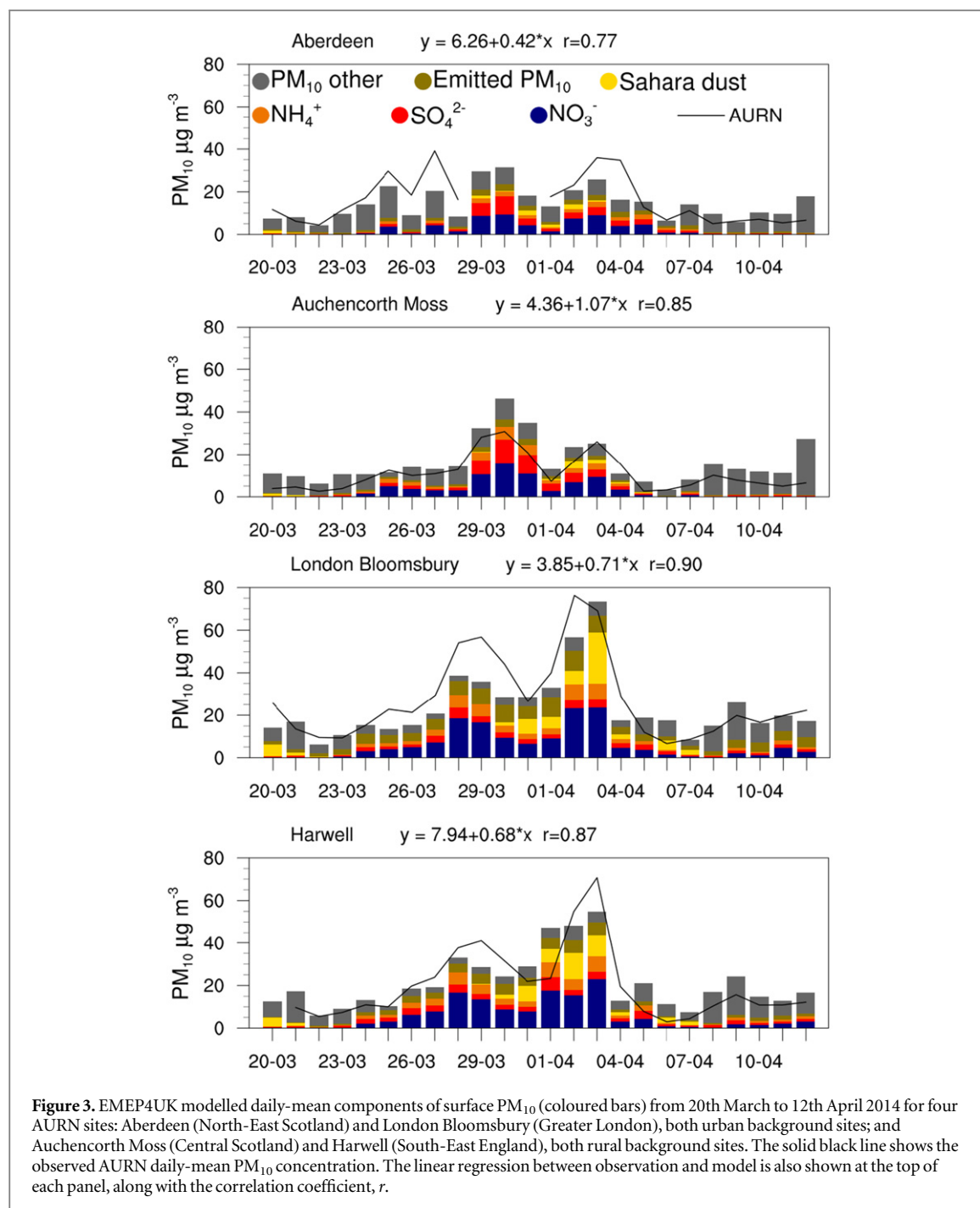
**Figure 2.** EMEP4UK modelled daily-mean components of surface $PM_{2.5}$ (coloured bars) from 20th March to 12th April 2014 for four AURN sites: Aberdeen (North-East Scotland) and London Bloomsbury (Greater London), both urban background sites; and Auchencorth Moss (Central Scotland) and Harwell (South-East England), both rural background sites. The solid black lines show the observed AURN daily-mean $PM_{2.5}$ concentrations. The linear regression between observation and model is shown at the top of each panel, along with the correlation coefficient, *r*.

shown in figures 4 and 5 for $PM_{2.5}$ and $PM_{10}$, respectively.

Figures 2 and 3 show that during the full period presented, $PM_{2.5}$ and $PM_{10}$ were dominated by secondary inorganic PM components, especially for $PM_{2.5}$. The estimated proportions of daily-mean $PM_{2.5}$ and $PM_{10}$ derived from UK emissions are shown in figures 6(a) and 7(a), respectively. The proportions were typically less than 40% from which it can be inferred that EP1 and EP2 both have a large component of PM derived from long-range transport

of secondary inorganic PM and secondary PM precursors. In contrast, on the 1st April, a date between the two episodes, up to 70% of $PM_{2.5}$ and $PM_{10}$ were estimated to have been generated from UK emissions. The contribution of transboundary transport to PM pollution events in the UK has been reported before; for example, Vieno *et al* (2014) showed that up to 80% of particle $NO_3^-$ in episodes may be imported into the UK from continental Europe.
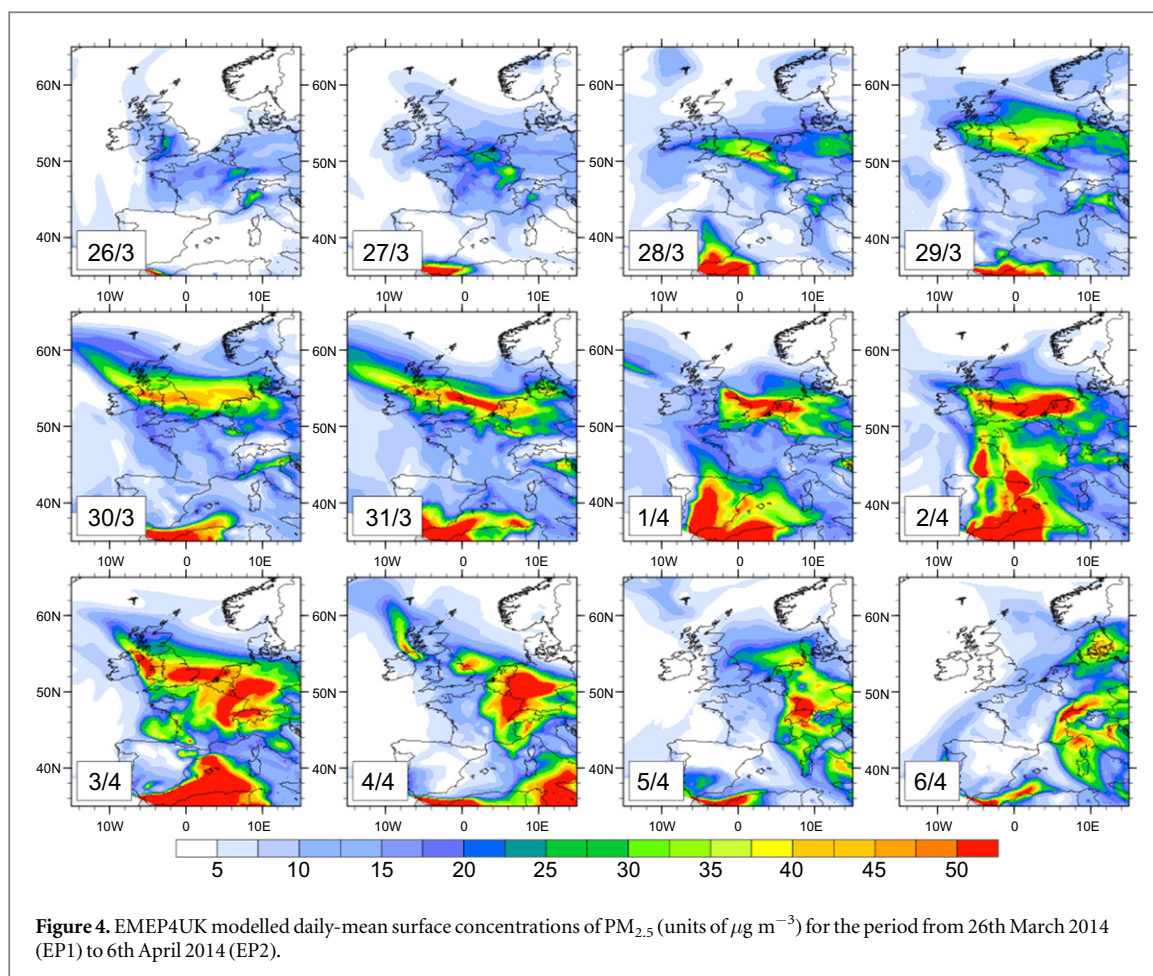
To further test the model, figure 8 shows the hourly modelled inorganic composition of $PM_{10}$ at

**Figure 3.** EMEP4UK modelled daily-mean components of surface $PM_{10}$ (coloured bars) from 20th March to 12th April 2014 for four AURN sites: Aberdeen (North-East Scotland) and London Bloomsbury (Greater London), both urban background sites; and Auchencorth Moss (Central Scotland) and Harwell (South-East England), both rural background sites. The solid black line shows the observed AURN daily-mean $PM_{10}$ concentration. The linear regression between observation and model is also shown at the top of each panel, along with the correlation coefficient, *r*.

Auchencorth Moss and at Harwell over the full time period compared against the MARGA measurements (uk-air.defra.gov.uk). Both the speciated MARGA measurements and the model follow the TEOM-FDMS $PM_{10}$ temporal variation shown in figure 3 relatively well. Gaps in the measurements indicate when the MARGA instrument was offline. As for total daily $PM_{2.5}$ and $PM_{10}$, the hourly observations of speciated $PM_{10}$ components during EP1 and EP2 are well captured by the model, both in timing and magnitude. The notable enhancement of $NO_3^-$ concentrations during this event is clearly evident in both the MARGA observations and in the model simulations as shown in figure 8. The fact that the $NO_3^-$ enhancement was

accompanied by an equivalent enhancement in $NH_4^+$ supports the conclusion that observed $PM_{2.5}$ enhancements are minimally impacted by dust or by $NO_3^-$ formation on dust surfaces.
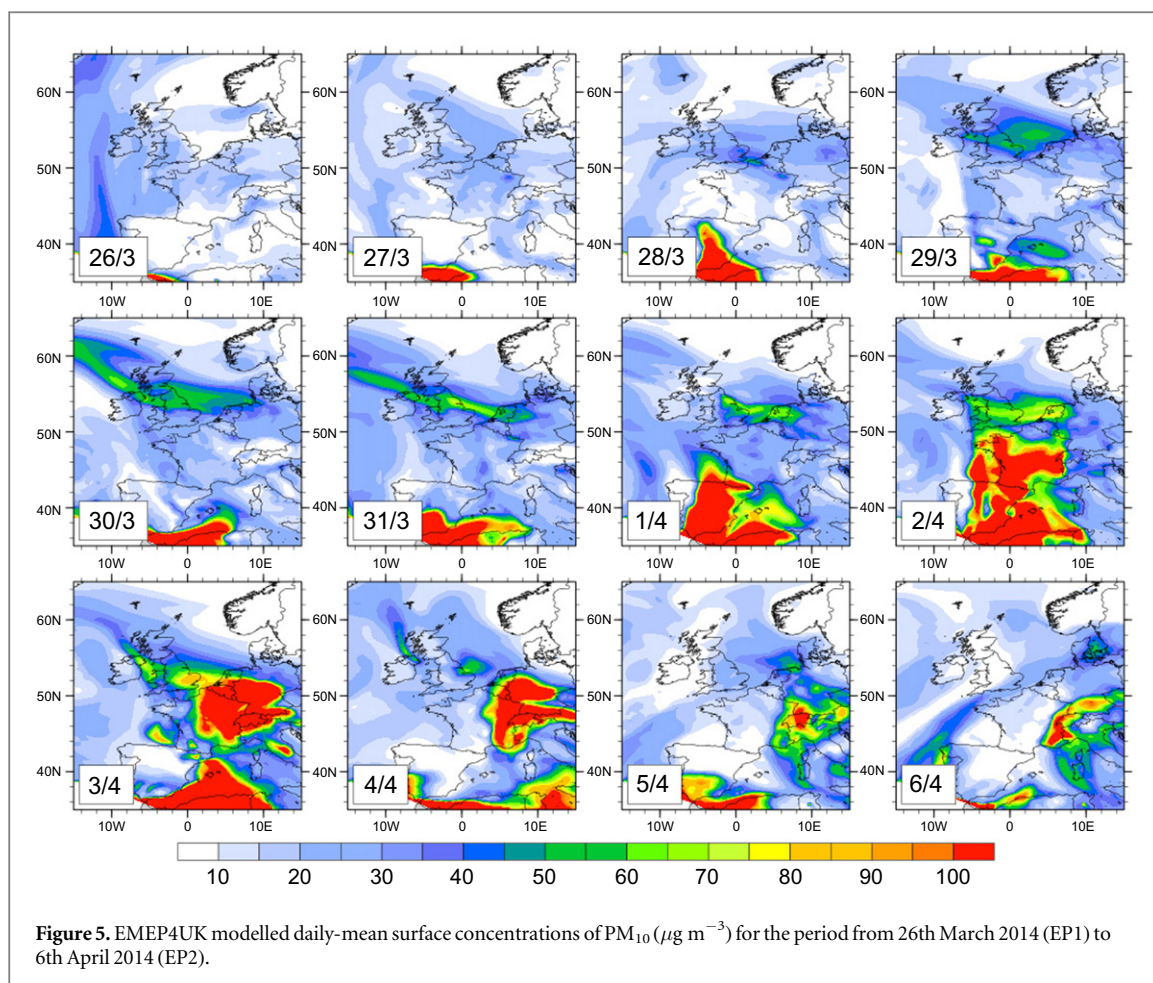
With respect to the Saharan dust component, the modelling indicates that the $PM_{2.5}$ and $PM_{10}$ composition differed between EP1 and EP2. Figures 6(b) and 7(b) show that Saharan dust constituted a greater component of both surface $PM_{2.5}$ and $PM_{10}$ during EP2 than during EP1. However, the dust was present at substantial surface concentrations only in the southern UK, as illustrated by the data presented here for the representative southern UK monitoring sites at Harwell and London Bloomsbury. Overall, the

**Figure 4.** EMEP4UK modelled daily-mean surface concentrations of $PM_{2.5}$ (units of $\mu g\ m^{-3}$) for the period from 26th March 2014 (EP1) to 6th April 2014 (EP2).

percentage of modelled Saharan dust in $PM_{2.5}$ and $PM_{10}$ is less than 20% for most of the UK for both EP1 and EP2. The geographic extent of the Saharan dust incursion to the surface (relative dust contribution to PM > 20%) for EP2 is shown in figures 6(b) and 7(b) for $PM_{2.5}$ and $PM_{10}$, respectively. During EP1 the high concentrations of PM in the plume were derived from anthropogenic emissions in continental Europe and advected to the UK as shown in figures 4 and 5. It was only during EP2 that the Saharan dust plume was also present at the surface (for parts of the UK) augmenting the secondary PM from continental Europe. The Saharan dust plume can be observed having evolved and moving north across Europe from the 30th March, arriving at the UK surface level on the 3rd of April (figure 2). Saharan dust in the atmosphere was also predicted by the model during EP1, but at higher altitudes with little impact at the surface. To visualise this altitudinal variation in PM composition between the two episodes, a 3D iso-surface of $\sim 5\ \mu g\ m^{-3}$ at the surface for $NO_3^-$ (green) and Saharan dust (yellow) is shown for 12:00 on 30th March during EP1 (figure 9(b)) and for 3rd April during EP2 (figure 10(b)). Figures 9(a) and 10(a) illustrate the geographic extent of when Saharan dust and $NO_3^-$ concentrations at the surface were greater than $1\ \mu g\ m^{-3}$ for these two occasions. Figure 9 shows that during

EP1 there was a low-altitude $NO_3^-$ plume advected in from continental Europe and a separate high altitude plume of Saharan dust. The model simulation shows that during EP1 the high altitude Saharan dust did not reach the UK surface apart from a small area in Wales as shown in figure 9(a). In contrast, figure 10 shows that for EP2 the Saharan dust was present at lower altitudes and had reached the surface in the south of the UK on the 3rd of April (the dust plume had also reached the surface in the UK on 2nd April).

From the analysis of these two PM events, which although temporally adjacent have distinct characteristics due to their air mass history, it has been shown that speciation measurements and modelling can allow the clear, quantitative diagnosis of the contributions to PM mass. In this case, the MARGA measurements characterised the inorganic composition and the modelling demonstrated how that composition developed. These measurements do not provide explicit information about the external/internal mixing properties of the PM, e.g. interactions of the secondary PM on the surfaces of mineral dust, which is likely to be occurring as noted in previous studies (Fairlie *et al* 2010). However, since in the EMEP4UK model the production of $NH_4NO_3$ is not linked to the additional particle surface area provided by the dust, the model's ability to reproduce measured concentration

**Figure 5.** EMEP4UK modelled daily-mean surface concentrations of $PM_{10}$ ($\mu g\ m^{-3}$) for the period from 26th March 2014 (EP1) to 6th April 2014 (EP2).

of these compounds suggests that the importance of the Saharan dust promoting secondary aerosol formation is not a key process for this event.

The modelled low levels of Saharan dust in EP1 (<20% of PM mass) coupled with the PM measurements indicate that the UK did not experience the consequences of a Saharan dust episode at that time. Rather, this was mainly an anthropogenic pollution event, exacerbated by the transport of secondary inorganic aerosol precursors emitted outside of the UK, which, in principle, could be mitigated against in future with appropriate international policies to reduce primary air pollutant emissions (especially $NO_x$ and $NH_3$). The importance of long-range transport of ammonium nitrate demonstrated here for this UK $PM_{2.5}$ episode corroborates previous work (Vieno *et al* 2014) and confirms similar findings on a European scale presented by Kiesewetter *et al* (2015). The policy implications derived from accurate identification of emission sources contributing to PM concentrations are discussed further in the next section.

## 4. Policy implications

The findings presented in this paper illustrate the capability of advanced ACTMs to reliably identify the underlying causes of complex air pollution episodes. The validation of the model results against chemically-speciated measurements of PM components further highlights the need for the development and implementation of better integrated model and measurement systems to routinely apportion sources and target the most relevant contributing activities for the design of effective pollution control policies (Reis *et al* 2015). It is also clear that it is feasible to provide robust information on the state of atmospheric composition as an evidence base to policy makers in a more operational manner.

The substantial contribution of long-range transport to ambient concentrations of fine PM in the UK, and the accurate identification of the relevant sources, has far-reaching policy implications. The pollution episode in spring 2014 was widely attributed to Saharan dust in the UK media, thus placing a (false) emphasis on a natural phenomenon, which cannot be addressed by policy action. The focus on an 'exciting headline' may be attributable to the complexity of communicating atmospheric processes and the public observations of dust-fall on surfaces. This suggests that while scientific analyses to determine the real causes of particular PM events through measurements and modelling are well established, the translation into communicating outside the atmospheric science arena
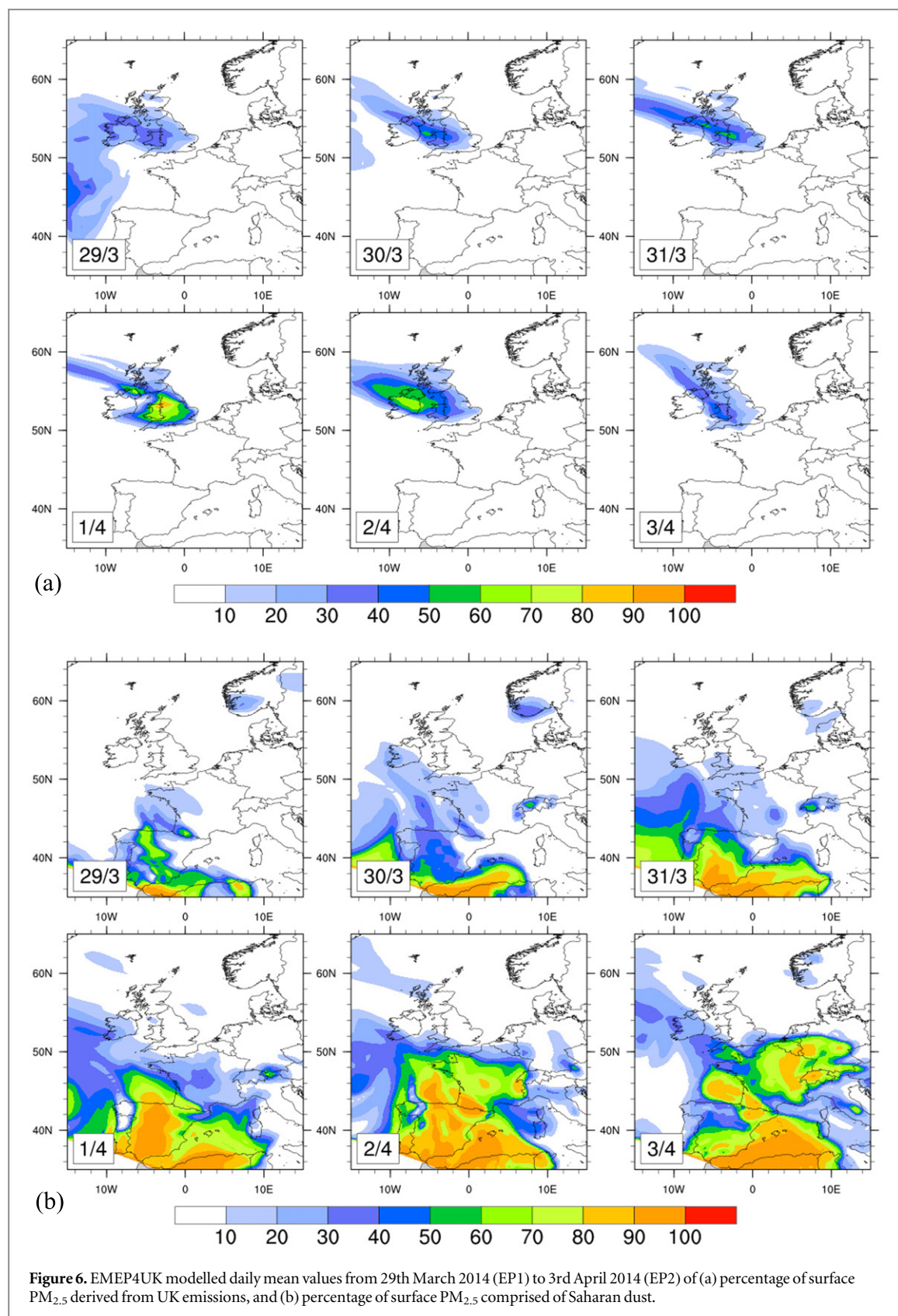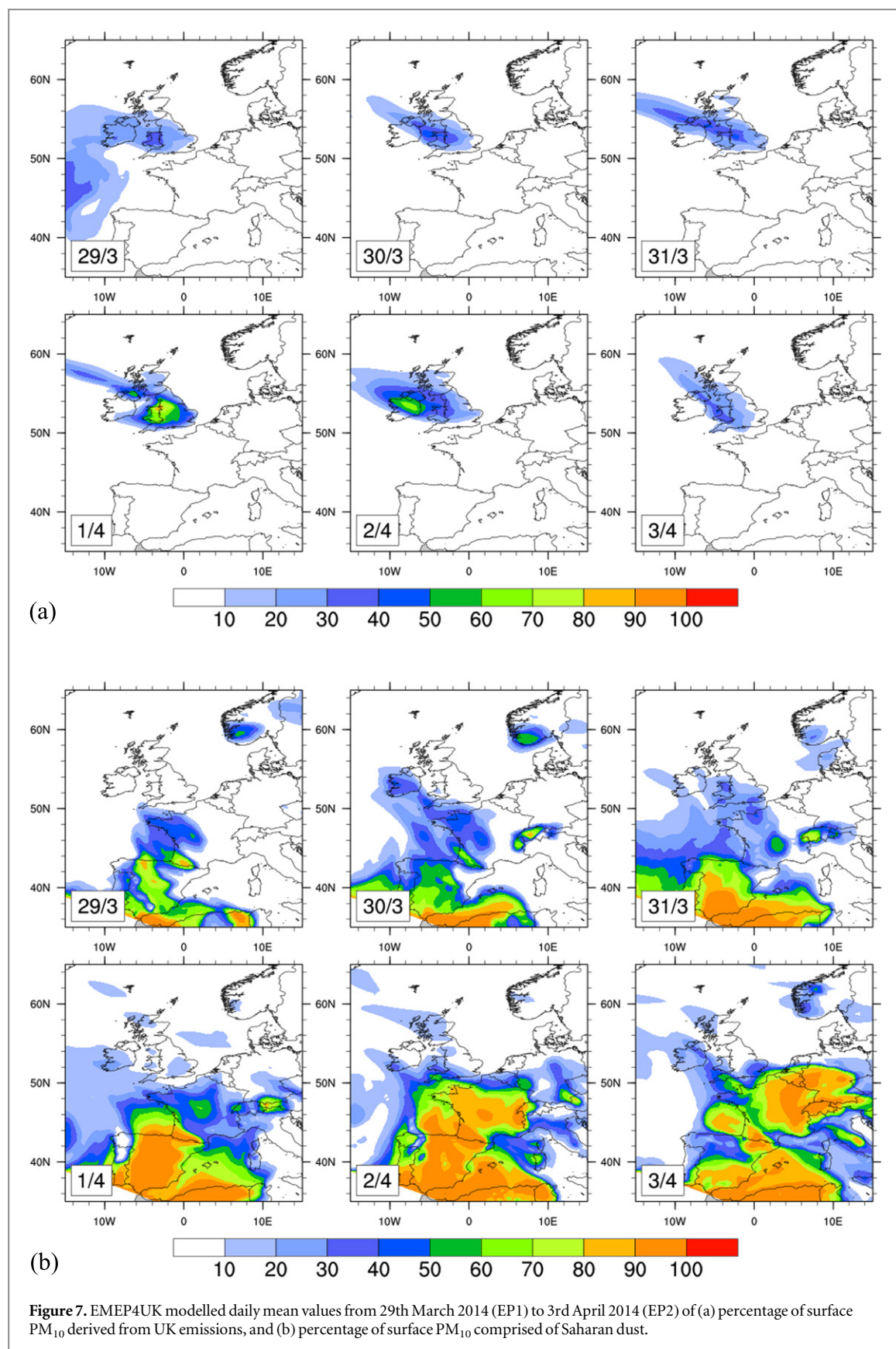
**Figure 6.** EMEP4UK modelled daily mean values from 29th March 2014 (EP1) to 3rd April 2014 (EP2) of (a) percentage of surface PM$_{2.5}$ derived from UK emissions, and (b) percentage of surface PM$_{2.5}$ comprised of Saharan dust.

needs improvement. Therefore, there is a danger that important messages to high-level policy-makers may be obscured by attention-catching headlines.

The initial mischaracterisation of the PM event may also represent a missed opportunity to inform and educate the general public about the role of anthropogenic emissions, specifically agricultural emissions of ammonia, as a key contributor to many high PM pollution events in recent years. As a consequence, the lack of public awareness may result in reduced public interest in, and a weaker policy mandate for, any emissions reduction targets for ammonia emissions and their contribution to transboundary fluxes of secondary aerosol components. The

**Figure 7.** EMEP4UK modelled daily mean values from 29th March 2014 (EP1) to 3rd April 2014 (EP2) of (a) percentage of surface $PM_{10}$ derived from UK emissions, and (b) percentage of surface $PM_{10}$ comprised of Saharan dust.

importance of public support has been illustrated by Reis *et al* (2012) specifically for the implementation of air pollution control policies through a robust science-policy interface. The extent to which explicit value judgements are a further underlying cause for the—to date—only moderate ambition levels in reducing ammonia emissions on a European scale (Voinov *et al* 2014) is difficult to quantify.
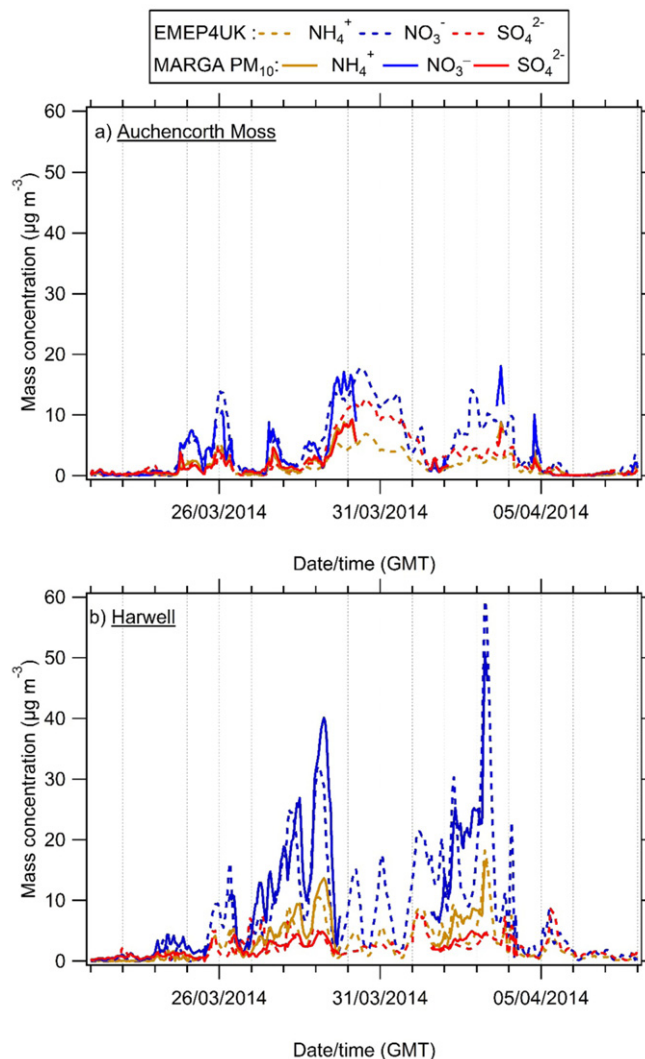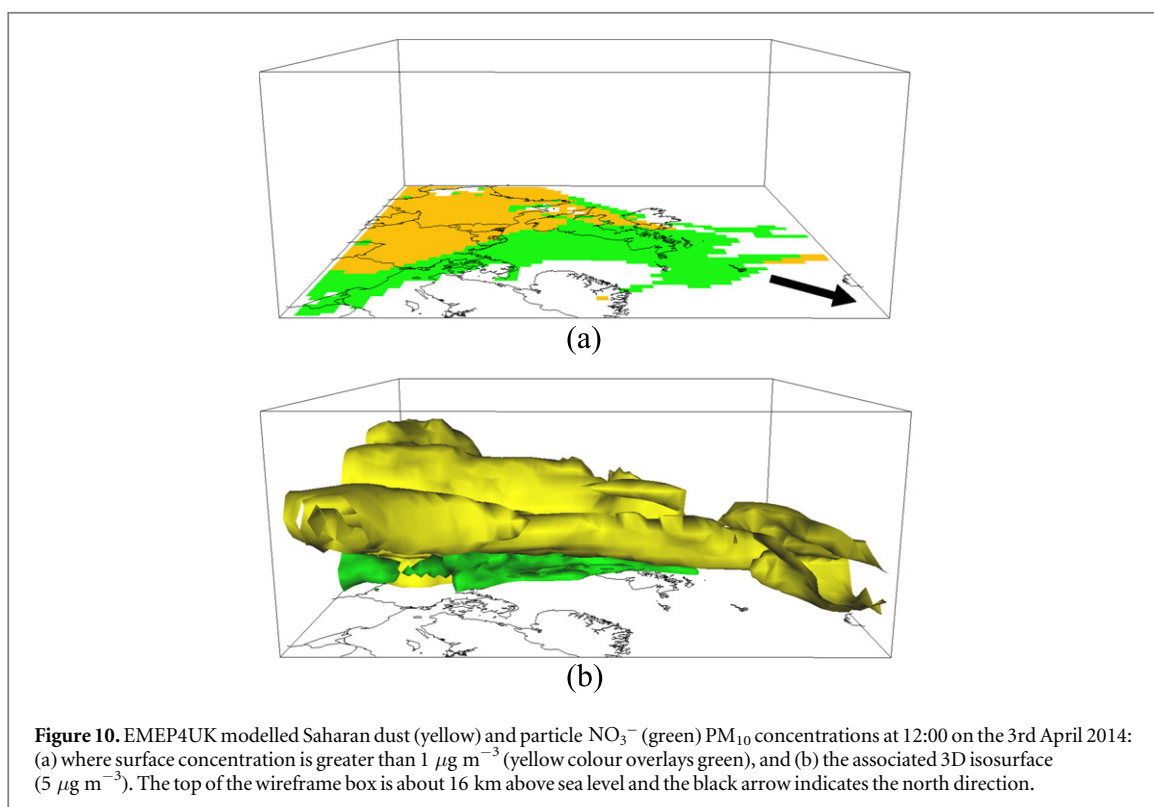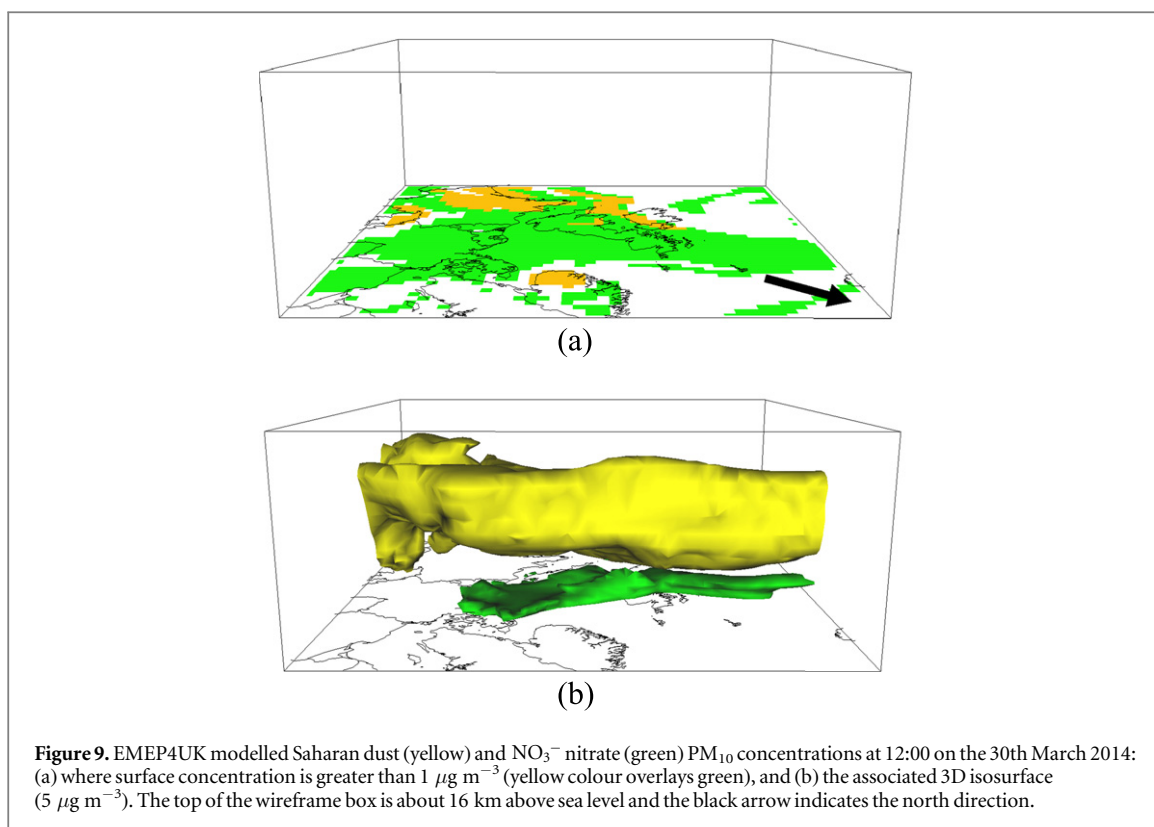
**Figure 8.** Hourly means calculated by the EMEP4UK model (dashed lines) and measured by the MARGA instruments (solid lines) at Auchencorth Moss (top) and Harwell (bottom) for the major secondary inorganic components of $PM_{10}$: $NH_4^+$ orange lines, $NO_3^-$ blue lines and $SO_4^{2-}$ red lines.

These implications are not only relevant for Europe. The PM pollution episodes in China and other fast growing economies (e.g. Oh *et al* 2015, Yan *et al* 2015, Zhang *et al* 2015, Zhou *et al* 2015, Tan *et al* 2016) are more severe examples of those in the UK, Paris, and other parts of Europe (Gualtieri *et al* 2015). While a major part of urban $PM_{2.5}$ in these other regions arises from primary $PM_{2.5}$ emissions from local transport and industrial activities, as has also been demonstrated for the UK (Vieno *et al* 2016), agricultural ammonia emissions have also been identified as key contributors. In these regions also, as for Europe, the need for close collaboration in policy development across provincial or national borders is evident and needs robust modelling and monitoring activities to provide underpinning scientific evidence. Such transboundary collaboration is emerging, e.g. in the case of tackling air quality and ozone pollution in Hong Kong and the Pearl River Delta (Zhong *et al* 2013). The combination of local measures and regional policies to reduce emissions within and far away from urban areas will likely be more effective in reducing the public health impacts of severe episodes of PM.

## 5. Conclusions

The EMEP4UK regional ACTM has been shown to successfully reproduce elevated surface PM concentrations during the spring of 2014 in the UK. The combined analysis of model results and speciated PM measurements indicates that these elevated PM concentrations were mainly driven by ammonium nitrate, much of which was derived from European $NO_x$ and $NH_3$ emissions outside the UK. The contribution of Saharan dust, which was widely reported in the UK media, has been shown to be restricted (at surface level) to the latter part of the elevated PM episode and regionally to the southern UK. Both types of PM and PM precursors arrived in the UK with the same southerly air flow, but originated from quite independent sources. The analyses presented in this paper

**Figure 9.** EMEP4UK modelled Saharan dust (yellow) and $NO_3^-$ nitrate (green) $PM_{10}$ concentrations at 12:00 on the 30th March 2014: (a) where surface concentration is greater than 1 $\mu$g m$^{-3}$ (yellow colour overlays green), and (b) the associated 3D isosurface (5 $\mu$g m$^{-3}$). The top of the wireframe box is about 16 km above sea level and the black arrow indicates the north direction.



**Figure 10.** EMEP4UK modelled Saharan dust (yellow) and particle $NO_3^-$ (green) $PM_{10}$ concentrations at 12:00 on the 3rd April 2014: (a) where surface concentration is greater than 1 $\mu$g m$^{-3}$ (yellow colour overlays green), and (b) the associated 3D isosurface (5 $\mu$g m$^{-3}$). The top of the wireframe box is about 16 km above sea level and the black arrow indicates the north direction.

illustrate the complementary use of high resolution observations (temporal and chemical speciation) and an advanced ACTM to identify the underlying causes of complex PM air pollution episodes. Specifically, the findings suggest that more emphasis on reducing ammonia emissions in Europe would have marked benefits in reducing episodic $PM_{2.5}$ concentrations and population exposure in the UK. The need for close collaboration in policy development across provincial or national borders applies worldwide.

## Acknowledgments

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