



The role of  
long-range transport  
and domestic  
emissions

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# The role of long-range transport and domestic emissions in determining atmospheric secondary inorganic particle concentrations across the UK

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## Abstract

Surface concentrations of secondary inorganic particle components over the UK have been analysed for 2001–2010 using the EMEP4UK regional atmospheric chemistry transport model. In early 2003 an episode of substantially elevated surface concentrations of ammonium nitrate was measured across the UK by the AGANET network. The EMEP4UK model was able accurately to represent both the long-term decadal surface concentrations and the episode in 2003. The latter was identified as consisting of three separate episodes, each of less than 1 month duration, in February, March and April. The primary cause of the elevated nitrate levels across the UK was meteorological, a persistent high pressure system, but whose varying location impacted the relative importance of transboundary vs. domestic emissions. Whilst long-range transport dominated the elevated nitrate in February, in contrast it was domestic emissions that mainly contributed to the March episode, and for the April episode both domestic emissions and long-range transport contributed. A prolonged episode such as the one in early 2003 can have substantial impact on annual average concentrations. The episode led to annual concentration differences at the regional scale of similar magnitude to those driven by long-term changes in precursor emissions over the full decade investigated here. The results demonstrate that a substantial part of the UK, particularly the south and south-east, may be close to or actually exceeding annual mean limit values because of import of inorganic aerosol components from continental Europe under specific conditions. The results reinforce the importance of employing multiple year simulations in the assessment of emissions reduction scenarios on PM concentrations and the need for international agreements to address the transboundary component of air pollution.

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## 1 Introduction

Atmospheric particulate matter (PM) concentrations are governed by the transport, transformation and deposition of many chemical species. PM has a range of impacts including on climate through radiative forcing and on human health. Considering the health impacts alone, exposure to PM<sub>2.5</sub> (the size fraction of particles with an aerodynamic diameter  $\leq 2.5 \mu\text{m}$ ) has been estimated to contribute to an average loss of life expectancy of around 6–7 months for residents of the UK, with an associated economic cost of some £16 billion per annum (IGCB, 2007). EU legislation sets standards for ambient concentrations of PM, and now includes an obligation on individual member states to reduce population-weighted exposure to PM<sub>2.5</sub> by a specified percentage between 2010 and 2020 (Heal et al., 2012).

The complexity of ambient PM composition and formation, combined with the influence of meteorology on chemistry, dispersion and deposition, considerably complicates pinpointing the contributions of different chemical pollutant emission sources to ambient PM at specific locations (USEPA, 2009; AQEG, 2012). Consequently, it is a complicated process to formulate cost-effective policy action to reduce harm caused by PM. The inorganic chemical components of PM, ammonium ( $\text{NH}_4^+$ ), sulphate ( $\text{SO}_4^{2-}$ ) and nitrate ( $\text{NO}_3^-$ ), constitute a major fraction of PM<sub>2.5</sub> (Putaud et al., 2010). The anthropogenic emissions of the gaseous precursors of inorganic PM – ammonia ( $\text{NH}_3$ ), sulphur dioxide ( $\text{SO}_2$ ) and nitrogen oxides ( $\text{NO}_x$ ) – are also subject to various legislation that seeks to limit and reduce either a country's total emissions or the emissions from individual sources or source sectors (Heal et al., 2012; Reis et al., 2012). For  $\text{SO}_2$  and  $\text{NO}_x$  in particular, emissions reductions have been very effective over the past few decades and this is reflected in reductions in ambient concentrations of the gases (ROTAP, 2012). Despite this, PM<sub>10</sub> concentrations across much of western Europe have not fallen significantly since the year 2000 (Harrison et al., 2008), as exemplified by recent analysis for sulphate.

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The longer lifetime of secondary PM components compared with their gaseous precursors means that transboundary transport and meteorology are important drivers. This is of particular relevance for the design of air quality policies seeking to reduce PM concentrations, especially as some limit values may be sensitive to a small number of high concentration episodes rather than long-term average concentrations. This is particularly important for the nitrate component which has shown to be the dominant component on days when  $PM_{10}$  exceeds  $50 \mu g m^{-3}$  (Yin and Harrison, 2008).

There remains a gap in understanding the extent to which domestic emissions and transboundary import of secondary inorganic PM contribute inter-annually and to episodes of elevated concentrations in the UK (RoTAP). This was the motivation for this work. Ambient concentrations of the inorganic components have been measured since the 1990s on a monthly average basis, as part of the UK Acid Gas and Aerosol Network (AGANET <http://uk-air.defra.gov.uk/networks/network-info?view=aganet>, see Tang et al., 2009, for description of the approach), providing a dataset against which to compare model output. In the present paper the EMEP4UK Eulerian atmospheric chemistry transport model (ACTM) (Vieno et al., 2009, 2010) is first evaluated against these measurements and then used in sensitivity simulations to assess the contributions of trans-boundary and domestic emissions to secondary inorganic particle concentrations in the UK and their inter-annual variability.

## 2 Methods

### 2.1 Model description and set-up

The EMEP4UK model used for this work is a nested regional ACTM based on version v3.7 of the main EMEP model (Simpson et al., 2012). A detailed description of the EMEP4UK model framework and setup are given in Vieno et al. (2010) and only brief relevant details are presented here.



turbation experiment was carried out by setting UK land emissions to zero for the year. This provides an approximate model estimate of the contribution of non-UK emissions to gaseous and particulate matter concentrations in the UK.

## 2.3 Measurement data

5 Model surface concentrations were compared with observational data from the UK Acid Gases and Aerosols Network (AGANet), which is one of the four component UK Eutrophying and Acidifying Pollutants monitoring networks (Conolly et al., 2011). The AGANet monitoring sites were established in 1999 for the long-term simultaneous measurement of the concentrations of  $\text{SO}_2$ ,  $\text{HNO}_3$  and  $\text{NH}_3$  gases and particle  $\text{NO}_3^-$ ,  $\text{SO}_4^{2-}$  and  $\text{NH}_4^+$ , in relation to changes in European emissions of  $\text{SO}_2$ ,  $\text{NO}_x$  and  $\text{NH}_3$ . Measurements are made using DELTA samplers (DENuder for Long-Term Atmospheric sampling) and have monthly-averaged time resolution (Sutton et al., 2001). The size cut-off of the DELTA sampler has been estimated to be  $\sim 4.5 \mu\text{m}$  (Tang et al., 2009), therefore the measured concentrations are between the  $\text{PM}_{2.5}$  and  $\text{PM}_{10}$  size fractions.

15 The EMEP4UK model assigns all  $\text{SO}_4^{2-}$  and  $\text{NH}_4^+$  components to  $\text{PM}_{2.5}$ . Modelled  $\text{NO}_3^-$  is assigned to both  $\text{PM}_{2.5}$  and  $\text{PM}_{\text{coarse}}$  which leads to potential negative bias in modelled vs. measured concentrations for  $\text{NO}_3^-$ . Four sites representing different areas of the UK (marked on Fig. 1) have been selected for the comparisons presented here: Strathvaich Dam (north-west Scotland); Bush (central Scotland); Rothamsted (south-east England); and Yarner Wood (south-west England).

## 3 Results

The time series of the modelled and observed monthly mean surface concentrations of particle  $\text{NO}_3^-$  and  $\text{SO}_4^{2-}$  at the four selected AGANet sites are shown in Fig. 2 and Fig. 3, respectively. The observations span the period 2001–2010, whereas the model values span the period 2001–2010. A peak in 2003 in  $\text{NO}_3^-$  concentrations can be seen

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in both observations and the EMEP4UK simulations. Figures 2 and 3, show generally good agreement between the two datasets for three of the four sites included here, as illustrated quantitatively by the correlation and linear regression statistics (Table 1). The model-measurement comparison at the Strathvaich Dam site is adversely impacted by two extreme measurement values in 2006 and 2007 not evident in the simulations.

The spatial pattern across the British Isles and the inter-year variability of modelled annual mean surface concentrations are shown in Fig. 4 for  $\text{NO}_2$ ,  $\text{SO}_2$  and  $\text{NH}_3$  and in Fig. 5 for particle  $\text{NO}_3^-$ ,  $\text{SO}_4^{2-}$  and  $\text{NH}_4^+$ . In each case, the top left panel shows the spatial distribution for 2001 and the other 9 panels show the differences in annual mean concentrations for each of years 2002–2010 relative to 2001. The maps for 2001 show highest concentrations of  $\text{NO}_2$  and  $\text{SO}_2$  over central and south east England, related to UK emission sources, and over the English Channel, mostly related to shipping emission sources. The highest concentrations of  $\text{NO}_2$  and  $\text{SO}_2$  over the UK are in 2003, with the lowest concentrations during 2008–2010. The extended periods of elevated  $\text{NO}_3^-$  during February to April 2003 were sufficient to enhance the annual average  $\text{NO}_3^-$  concentration across the whole of the UK in 2003 by between 0.2–0.3  $\mu\text{g N m}^{-3}$  compared with preceding and subsequent years (Fig. 5a), with a even larger enhancement in the annual mean for 2003 of 0.2–0.5  $\mu\text{g N m}^{-3}$  for  $\text{NH}_4^+$  (Fig. 5c). In contrast, the somewhat less elevated  $\text{SO}_4^{2-}$  concentrations during this period led to a modest increase in annual average  $\text{SO}_4^{2-}$  for 2003 of 0.0–0.1  $\mu\text{g S m}^{-3}$  (Fig. 5b). The spatial distribution of  $\text{NH}_3$  shows a very different pattern to the other modelled components, with highest modelled concentrations in Brittany and north-west France and north-west England, reflecting the distribution of modelled  $\text{NH}_3$  emissions which mainly arise from agricultural sources.

The concentrations of the particle components  $\text{NO}_3^-$ ,  $\text{SO}_4^{2-}$  and  $\text{NH}_4^+$  are spatially smoother across the UK than the gaseous precursors (Figs. 4 and 5). The modelled annual surface concentrations of  $\text{NO}_2$  and  $\text{SO}_2$  (Fig. 4a and b) show that the concentrations of these gaseous components decline during 2001–2010 by substantially more than the decline in  $\text{NO}_3^-$  and  $\text{SO}_4^{2-}$ . Over much of the UK (particularly England),

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declines in modelled  $\text{NO}_2$  and  $\text{SO}_2$  between 2001 and 2010 exceed  $1 \mu\text{gN}/\text{Sm}^{-3}$  in 2010 compared with the  $0.1\text{--}0.2 \mu\text{gNm}^{-3}$  decline in  $\text{NO}_3^-$ , and the  $0.1\text{--}0.3 \mu\text{gSm}^{-3}$  decline in  $\text{SO}_4^{2-}$  (up to  $0.4 \mu\text{gSm}^{-3}$  decline in eastern England). On the other hand the model shows concentrations of  $\text{NH}_3$  hardly changing over the decade; in fact increasing slightly, up to  $\sim 0.2 \mu\text{gNm}^{-3}$  over England, especially for 2009 and 2010 (Fig. 4c), whereas, with the exception of 2003, the modelled concentration of  $\text{NH}_4^+$  in PM decreases from 2001–2010. Of note also, is a decrease of  $\text{SO}_2$  annual surface concentration over the North Sea from 2007 onwards (Fig. 4b).

Figure 6a and b (upper panels) show the modelled monthly-mean surface concentrations of  $\text{NO}_2$  and  $\text{SO}_2$ , respectively, for the first 5 months of 2003, which covers the period of high secondary inorganic particle concentrations shown in (Figs. 2 and 3). To highlight the role of UK sources, the differences between the base simulations and the simulations with zero UK emissions are shown in the lower panels of Fig. 6a and b, with the data expressed as the percentage of the modelled concentrations that are directly attributable to UK domestic emissions (i.e.  $100 \times (\text{Base Run} - \text{Experiment})/\text{Base}$ ), again as monthly averages. While the lower maps clearly show the dominating contribution of UK domestic sources to  $\text{NO}_2$  and  $\text{SO}_2$  concentrations over mainland UK, a smaller contribution in the vicinity of major shipping channels reflects the fact that the scenario treated international shipping as part of the non-UK emissions.

Figure 7a and b shows similar model results to Fig. 6 but for surface concentrations of particle  $\text{NO}_3^-$  and  $\text{SO}_4^{2-}$ , respectively. For these components, there is a smaller percentage contribution from UK sources than for  $\text{SO}_2$  and  $\text{NO}_x$  concentrations.

The highest concentrations of  $\text{NO}_2$  and  $\text{SO}_2$  occurred during February and March (Fig. 6), with highest concentrations for  $\text{NO}_3^-$  and  $\text{SO}_4^{2-}$  occurring during February, March and April (Fig. 7). Figure 7 shows that, for February, up to 40% of the monthly average  $\text{NO}_3^-$  concentrations over the UK are attributable to UK emissions. In March and April, the UK contribution to  $\text{NO}_3^-$  concentrations rises to up to 80%. The spatial pattern of the UK contribution to these concentrations differs between the months, with

February showing the smallest contribution of UK sources to  $\text{SO}_2$ ,  $\text{NO}_2$ ,  $\text{NO}_3^-$  and  $\text{SO}_4^{2-}$  concentrations. In contrast, the episodes in March and April 2003 have substantially larger contributions of UK emissions to  $\text{NO}_3^-$  and  $\text{SO}_4^{2-}$  concentrations.

A detailed comparison for 2003 between the measured and modelled  $\text{NO}_3^-$  concentrations at the Bush 1 site (Scotland) is shown in Fig. 8 with the modelled values presented as both monthly and daily means. There is a close agreement between observations (red line) and model (blue line) for these high concentration episodes, with the monthly values broadly agreeing within 10%.

The monthly modelled concentrations for the simulation with zero UK emissions are also shown in Fig. 8 (green line). The modelled monthly  $\text{NO}_3^-$  concentrations (blue line) were enhanced by  $2.6 \mu\text{g m}^{-3}$  by UK emissions in February, but by  $5.0 \mu\text{g m}^{-3}$  in March and by  $2.6 \mu\text{g m}^{-3}$  in April as compared with the model simulation with no-UK emissions (green line). The daily mean model  $\text{NO}_3^-$  concentrations highlight substantial temporal variability within this February–April period. The daily average surface concentrations (orange line of Fig. 8) show three separate episodes; the first approximately matches the period 12–28 February (F), the second 10–27 March (M) and the third 1–30 April (A).

The characteristic differences between these three periods are illustrated in Fig. 9. Here the 12:00 LT wind vector is superimposed to the mean modelled surface concentration of  $\text{PM NO}_3^-$  for selected days during the three component episodes. It is seen that 12–15 February (episode F) and 17–20 March (episode M) were associated with stagnant air masses allowing  $\text{NO}_3^-$  PM concentrations to build up, while the period 11–14 April (episode A) was associated with a highlight polluted air mass arriving from the East.

The UK February episode was associated with an easterly light wind advecting  $\text{PM NO}_3^-$  produced in the area of the north of France, Holland, north of Germany, and Denmark, where the centre of the high pressure was located (Fig. 9). During the March episode, the centre of the high pressure was over the UK with an associated light wind, clear sky, and cooler conditions leading to the accumulation of  $\text{NO}_3^-$  from UK emissions

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with little import of  $\text{NO}_3^-$  or its precursors from outside the UK. The April episode was a mixture of conditions described for February and March.

Based on the simulations it is possible to estimate the annual contribution of non-UK emissions to the different components of  $\text{PM}_{2.5}$  at the four study sites, Strathvaich Dam, Bush 1, Rothamsted and Yarner Wood. This is summarized Fig. 10 for the year 2003, also including the contribution of fine primary particulate matter ( $\text{PPM}_{2.5}$ ). Pollution import from non-UK sources ranges from an estimated 41 % for Bush 1, up to 62.5 % for Yarner Wood, highlighting the importance of transboundary pollution import on UK  $\text{PM}_{2.5}$  concentrations.

The same model results for 2003 can be expressed in terms of the contribution of non-UK emissions to the current European Commission (EC, 2013) limit value for  $\text{PM}_{2.5}$  and to the World Health Organisation (WHO, 2005) guideline value for  $\text{PM}_{2.5}$  at each of the four sites (Table 2). For these example sites, up to 18 % and 45 % of the limit and guideline values, respectively, is provided by non-UK emissions.

## 4 Discussion

Inorganic particle components were simulated over the period 2001–2010. The smoother distribution of particle components as compared to their gaseous precursors reflects the longer timescales for forming these secondary pollutants, as compared with the emissions-driven patterns for the primary pollutant gases (AQEG, 2012). The highest modelled concentrations over this period are in 2003, particularly for  $\text{PM}$   $\text{NO}_3^-$  and  $\text{NH}_4^+$ , and to a lesser extent for  $\text{SO}_4^{2-}$ , whilst lowest concentrations for each of these components are in 2008–2010.

The notably high  $\text{PM}$   $\text{NO}_3^-$  concentrations in February to April 2003 were observed at AGANet stations across the UK and could be well reproduced by the model (Fig. 2, Table 1). Concentrations of  $\text{PM}$   $\text{SO}_4^{2-}$  were also elevated during this period, although by a smaller amount, and were also well captured by the model (Fig. 3, Table 1). The magnitude of this elevation in annual average  $\text{PM}$   $\text{NO}_3^-$  concentration in 2003 is greater

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than the decline in annual average concentration across the whole decade to 2010 of  $0.1\text{--}0.2\ \mu\text{gNm}^{-3}$  (Fig. 5).

The simulated changes in the gaseous precursors for 2001–2010 follow the reductions in UK emissions over that period especially for  $\text{NO}_2$  and  $\text{SO}_2$  (MacCarthy et al., 2012). The change of  $\text{SO}_2$  annual surface concentration especially after 2007 over the North Sea (Fig. 4b) is a direct response to the introduction of a sulphur emission control area (SECA) in the North Sea, including the English Channel, by the 2007 MARPOL convention on marine pollution (Dore et al., 2007). Under the convention the sulphur content of bunker fuel was restricted to 1.5% by mass in 2007 (and will be further reduced to 0.1% in SECAs by 2020). This has resulted in a substantial reduction of emissions of  $\text{SO}_2$  from the shipping sector.

The results in Figs. 4 and 5 illustrate the non-linear relationship between changes over time in  $\text{SO}_2$  and  $\text{NO}_2$  surface concentrations over the 2001–2010 decade and changes in the respective PM  $\text{SO}_4^{2-}$  and  $\text{NO}_3^-$  concentrations. The sensitivity of PM  $\text{SO}_4^{2-}$  to changes in its precursors is, however, considerably greater than for  $\text{NO}_3^-$ . The small decline in  $\text{NO}_3^-$  and low sensitivity to UK  $\text{NO}_x$  emission found in this work was supported by the results in Harrison et al. (2013). The formation of both  $\text{NO}_3^-$  and  $\text{SO}_4^{2-}$  requires  $\text{NH}_4^+$  as a counter-ion and there appear to be sufficient  $\text{NH}_3$  emissions not to be a limiting factor to  $\text{SO}_4^{2-}$  formation. Conversely, UK  $\text{NO}_x$  emissions are still relatively high, especially in urban areas, so with an abundance of  $\text{NO}_x$  available for formation of ammonium nitrate available  $\text{NH}_3$  eventually may be consumed. Consequently, in areas of high  $\text{NO}_x$  emissions,  $\text{NO}_3^-$  formation appears to be more sensitive to  $\text{NH}_3$  emissions than is the case for  $\text{SO}_4^{2-}$  formation. This is consistent with Redington et al. (2009) whose modelling showed that  $\text{SO}_4^{2-}$  formation in the UK was less sensitive to a 30%  $\text{NH}_3$  emissions reduction than  $\text{NO}_3^-$  formation which only responded slightly to the same  $\text{NH}_3$  emissions reduction.

The modelled annual average  $\text{NH}_4^+$  shows a change between 2001 and 2010 over the UK which is intermediate between that of  $\text{NO}_3^-$  and  $\text{SO}_4^{2-}$  (Fig. 5c). By 2010,  $\text{NH}_4^+$

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concentrations decreased by 0.3–0.4  $\mu\text{g N m}^{-3}$  over most of England, but, as was the case for  $\text{NO}_3^-$  concentrations, annual average  $\text{NH}_4^+$  concentrations in 2003 were elevated by 0.2–0.3  $\mu\text{g N m}^{-3}$  compared with preceding and subsequent years. This confirms that the episodes of elevated  $\text{NO}_3^-$  in 2003 were driven by ammonium nitrate specifically. The modelled decrease in  $\text{PM NH}_4^+$  concentrations as compared with minimal decrease (and some increase) in  $\text{NH}_3$  concentrations over the period 2001–2010 is consistent with the conclusions of Bleeker et al. (2009) and Horvath et al. (2009) for other parts of Europe that reducing  $\text{SO}_2$  emissions have contributed to maintaining or even increasing gaseous  $\text{NH}_3$  concentrations.

The geographic origins of the PM episodes have been investigated in the model perturbation experiment. The monthly average surface concentrations for the zero UK emissions experiment show that surface concentrations of  $\text{SO}_2$  and  $\text{NO}_2$  are mainly driven by UK emissions (Fig. 6) and by similar proportions of UK emissions throughout the period of high surface concentrations of  $\text{NO}_3^-$ . However, the proportions of the  $\text{NO}_3^-$  that are derived from UK and non-UK emissions changes between months (Fig. 7). The model results show that for February 2003 trans-boundary emissions had a small influence on  $\text{NO}_3^-$ , whereas for March and April the trans-boundary transport of  $\text{NO}_3^-$  and/or its precursors was substantial. Abdalmogith et al. (2006) suggest that the annual average import of  $\text{NO}_3^-$  aerosol to the UK from Europe (as an average of 2002 and 2003) is between 35–65 % of the UK total  $\text{NO}_3^-$  concentration. Our study has found that, for 2003 (Fig. 7), the import to the UK from Europe was in the range 20–60 % of UK total  $\text{NO}_3^-$  concentrations, with this proportion varying between the three episodes (labelled F, M and A in Fig. 8). Abdalmogith et al. (2006) concluded that the 2003  $\text{NO}_3^-$  spring event was not well represented by their model, and the low emissions resolution (10 km  $\times$  10 km grid) was suggested as a possible cause. In the present study work the elevated  $\text{NO}_3^-$  concentrations are well represented by the EMEP4UK model at 5 km  $\times$  5 km resolution. However, we find that simulation at 50 km  $\times$  50 km horizontal spatial resolution of the EMEP4UK model outer domain also represented these

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annual variability of surface concentrations of nitrate for the 2001–2010 decade as a response to changes in meteorological conditions is larger than the effect of changes in anthropogenic emissions. For instance, up to 60 % of  $\text{NO}_3^-$  may be imported from outside the UK under specific conditions.

5 Our results highlight how inter-annual variability can profoundly affect the sensitivity to the attainment of limit values for ambient PM concentrations as a result of non-domestic contributions from transboundary air pollution transport.

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**Table 1.** Mean concentrations, and correlation and regression statistics, for monthly-averaged modelled and measured  $\text{NO}_3^-$  and  $\text{SO}_4^{2-}$  in particulate matter for the period 2001–2010 at four sites of the AGANet network: Strathvaich Dam (north-west Scotland), Bush 1 (central Scotland), Rothamsted (south-east England), and Yarner Wood (south-west England). The comparison is based on a linear fit where measurement = slope · model + intercept.

	Particulate $\text{NO}_3^-$			
	Strathvaich Dam	Bush 1	Rothamsted	Yarner Wood
Measurement mean	$0.49 \mu\text{g m}^{-3}$	$1.37 \mu\text{g m}^{-3}$	$3.35 \mu\text{g m}^{-3}$	$1.98 \mu\text{g m}^{-3}$
Model mean	$0.77 \mu\text{g m}^{-3}$	$1.42 \mu\text{g m}^{-3}$	$2.73 \mu\text{g m}^{-3}$	$2.23 \mu\text{g m}^{-3}$
<i>r</i>	0.49	0.91	0.81	0.86
Slope	0.59	0.96	0.68	0.95
Intercept	$0.48 \mu\text{g m}^{-3}$	$0.10 \mu\text{g m}^{-3}$	$0.44 \mu\text{g m}^{-3}$	$0.34 \mu\text{g m}^{-3}$

	Particulate $\text{SO}_4^{2-}$			
	Strathvaich Dam	Bush 1	Rothamsted	Yarner Wood
Measurement mean	$0.57 \mu\text{g m}^{-3}$	$0.94 \mu\text{g m}^{-3}$	$1.75 \mu\text{g m}^{-3}$	$1.20 \mu\text{g m}^{-3}$
Model mean	$0.61 \mu\text{g m}^{-3}$	$0.95 \mu\text{g m}^{-3}$	$1.48 \mu\text{g m}^{-3}$	$1.28 \mu\text{g m}^{-3}$
<i>r</i>	0.72	0.79	0.65	0.69
Slope	0.86	0.76	0.56	0.65
Intercept	$0.12 \mu\text{g m}^{-3}$	$0.24 \mu\text{g m}^{-3}$	$0.50 \mu\text{g m}^{-3}$	$0.36 \mu\text{g m}^{-3}$

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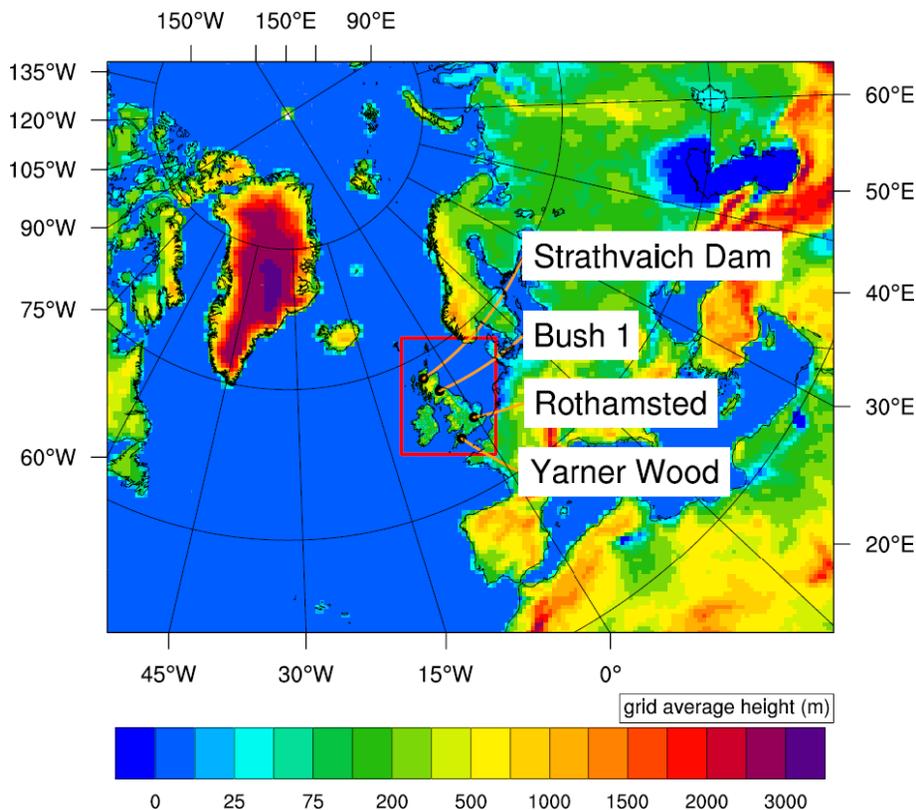
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**Table 2.** Model simulated contributions of Continental European  $\text{PM}_{2.5}$  import to the current European Commission limit value (EC, 2013) and to the World Health Organisation guideline value (WHO, 2005) at each of the four sites for the EMEP4UK model simulations for the year 2003.

Continental European contribution	Strathvaich Dam	Bush 1	Rothamsted	Yarner Wood
EC limit value of $25 \mu\text{g m}^{-3}$	5 %	8 %	18 %	15 %
WHO guideline of $10 \mu\text{g m}^{-3}$	14 %	20 %	45 %	38 %



**Fig. 1.** The EMEP4UK Greater European domain, modelled at  $50\text{km} \times 50\text{km}$  horizontal resolution and, outlined in red, the nested British Isles domain, modelled at  $5\text{km} \times 5\text{km}$  horizontal resolution. The colour scale indicates grid-average altitude. The four UK AGANet measurement sites used in this study are also shown.

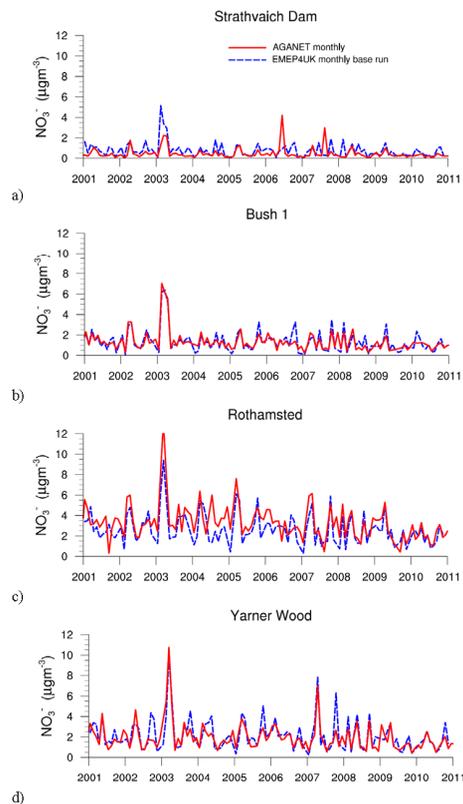
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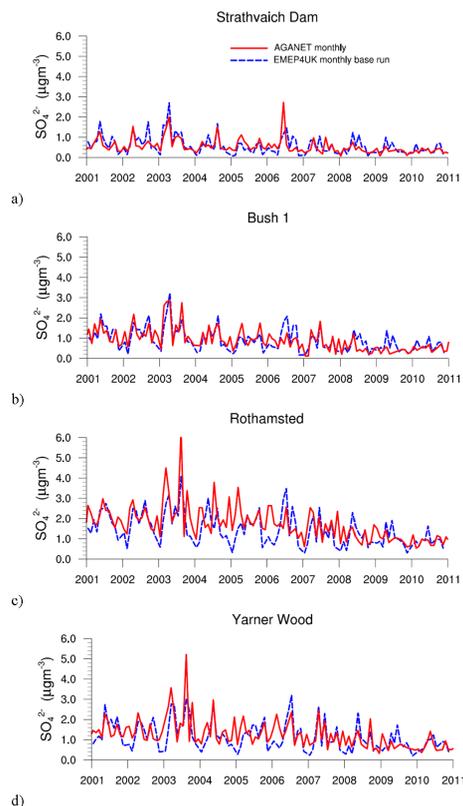
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**Fig. 2.** Monthly average surface concentrations of particulate matter nitrate, observed (RED) and modelled (BLUE), for 2001–2010 at four sites of the AGANet network: Strathvaich Dam (north-west Scotland), Bush (central Scotland), Rothamsted (south-east England), and Yarner Wood (south-west England).

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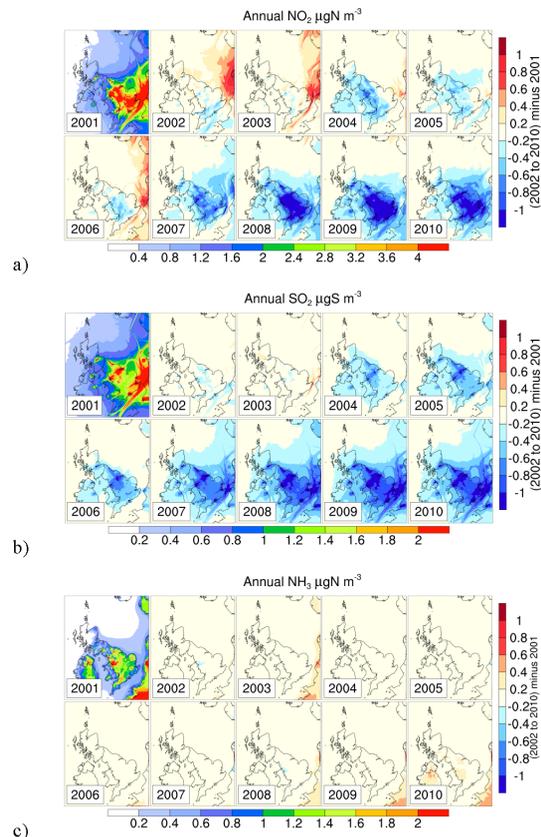
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**Fig. 3.** Monthly average surface concentration of particulate matter sulphate, observed (RED) and modelled (BLUE), for 2001–2010 at four sites of the AGANet network: Strathvaich Dam (north-west Scotland), Bush (central Scotland), Rothamsted (south-east England), and Yarner Wood (south-west England).

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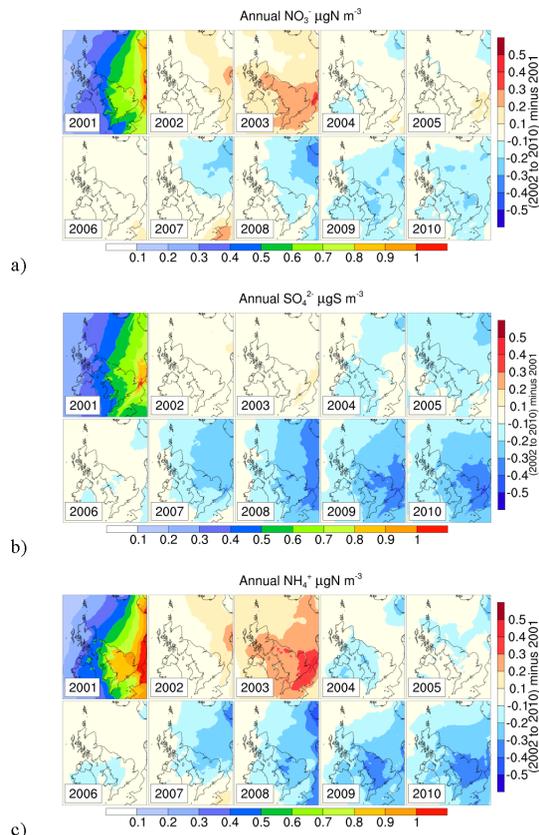
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**Fig. 4.** Modelled annual average surface concentrations of (a)  $\text{NO}_2$ , (b)  $\text{SO}_2$  and (c)  $\text{NH}_3$ . The top left panel of each figure shows the concentrations in 2001 (horizontal scales in  $\mu\text{gN m}^{-3}$  or  $\mu\text{gS m}^{-3}$ , as relevant). The remaining panels illustrate the difference in surface concentrations in each of years 2002 to 2010 compared with 2001 (vertical scales in  $\mu\text{gN m}^{-3}$  or  $\mu\text{gS m}^{-3}$ , as relevant).

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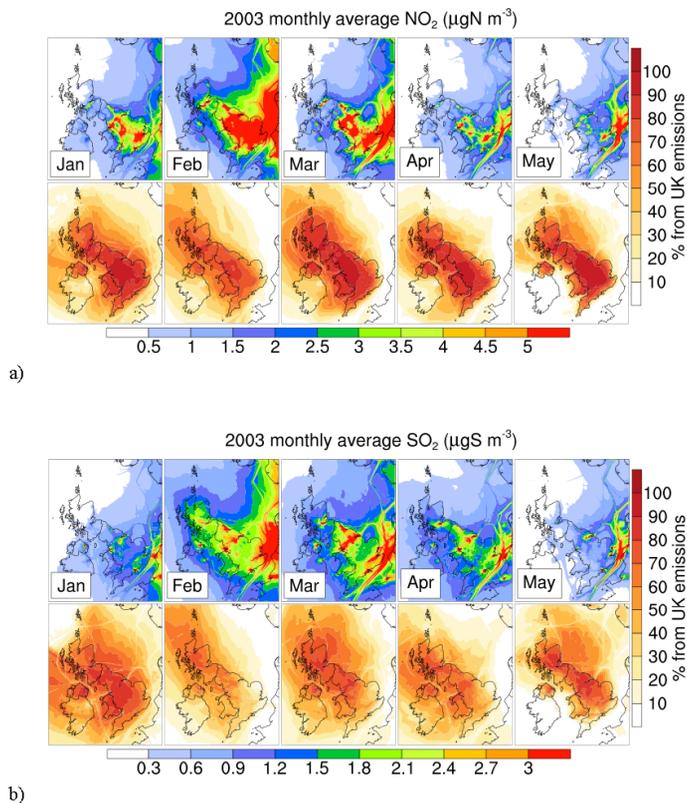
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**Fig. 5.** Modelled annual average surface concentrations of **(a)** NO<sub>3</sub><sup>-</sup>, **(b)** SO<sub>4</sub><sup>2-</sup> and **(c)** NH<sub>4</sub><sup>+</sup>. The top left panel of each figure shows the concentrations in 2001 (horizontal scales in µgN m<sup>-3</sup> or µgS m<sup>-3</sup>, as relevant). The remaining panels illustrate the difference in surface concentrations in each of years 2002 to 2010 compared with 2001 (vertical scales in µgN m<sup>-3</sup> or µgS m<sup>-3</sup>, as relevant).

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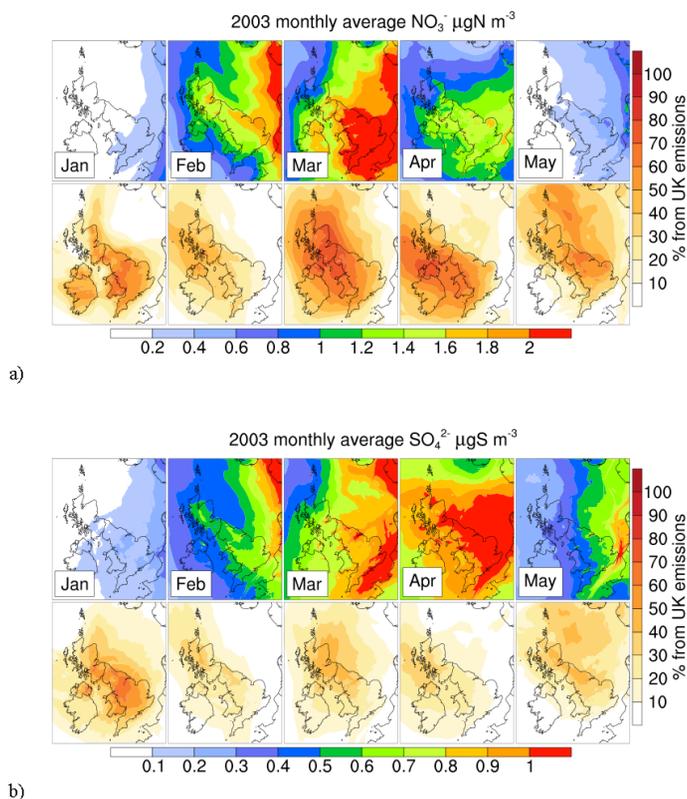
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**Fig. 6.** Modelled monthly average surface concentrations and UK contributions for **(a)**  $\text{NO}_2$  and **(b)**  $\text{SO}_2$  through January to May 2003. In each case the upper panel shows the mean concentrations ( $\mu\text{gN m}^{-3}$  or  $\mu\text{gS m}^{-3}$ ), while the lower panel shows the percentage contribution from UK emissions (calculated as  $[100 \times (\text{Base} - \text{no UK emi})/\text{Base}]$  between the base simulation and the simulation with zero UK emissions).

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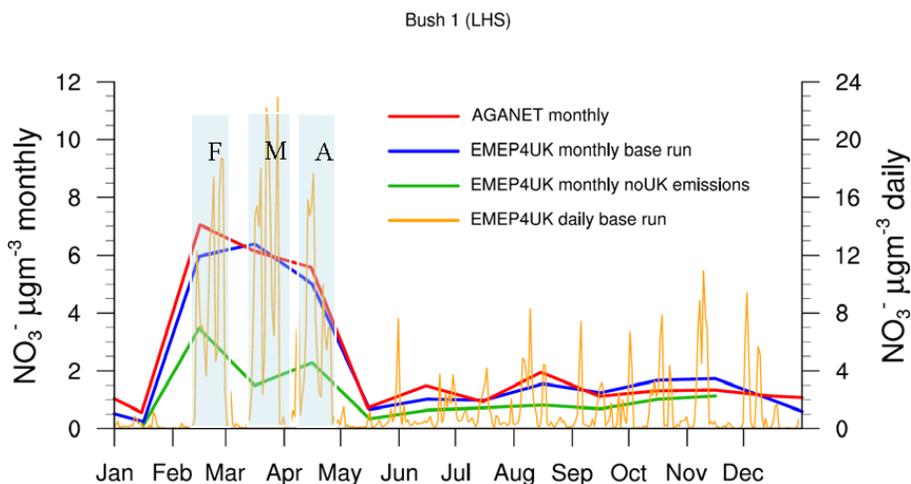
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**Fig. 7.** Modelled monthly average surface concentrations and UK contributions for **(a)**  $\text{NO}_3^-$  and **(b)**  $\text{SO}_4^{2-}$  through January to May 2003. In each case the upper panel shows the mean concentrations ( $\mu\text{gN m}^{-3}$  or  $\mu\text{gS m}^{-3}$ ), while the lower panel shows the percentage contribution from UK emissions (calculated as  $[100 \times (\text{Base} - \text{noUKemi})/\text{Base}]$  between the base simulation and the simulation with zero UK emissions).

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**Fig. 8.** Modelled and observed monthly mean (left axis) and daily mean (right axis) surface concentrations of  $\text{NO}_3^-$  at the Bush site (Scotland) for the year 2003. The red line is the measured monthly values from the AGANet network, the blue line is the modelled monthly means for the base run, the green line is the modelled monthly means for the experiment with no UK emissions, and the orange line is the modelled daily means for the base run. (All model values are averages of hourly data). The blue shadows highlight the three episodes labelled F, M and A.

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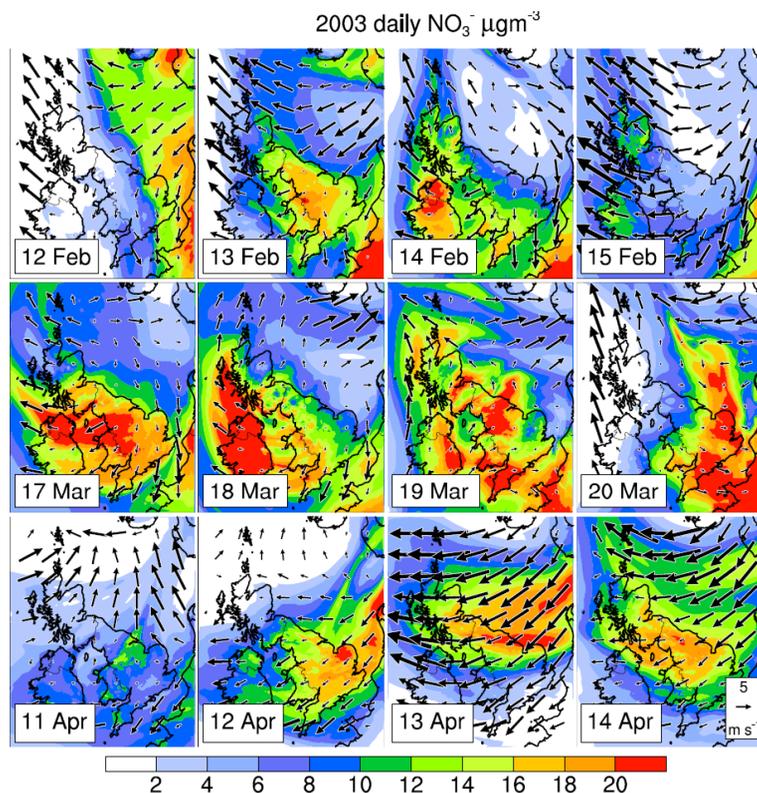
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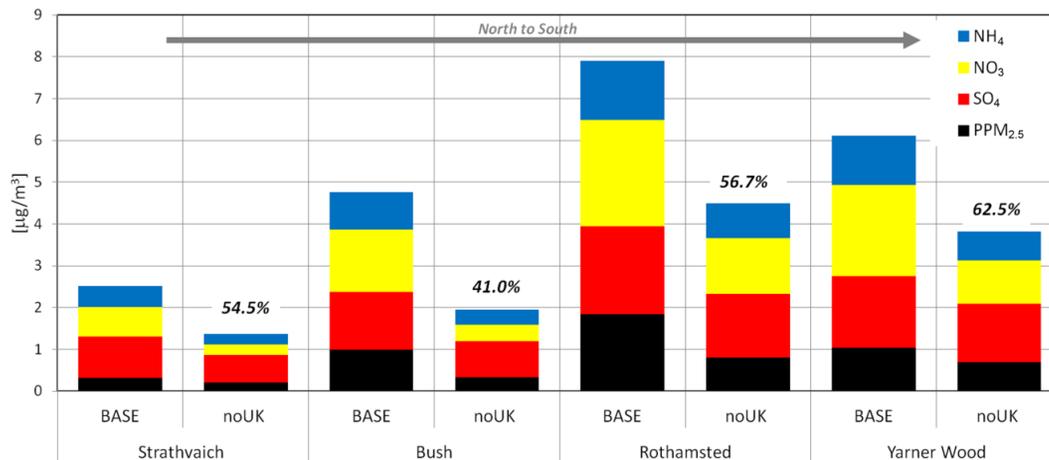
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**Fig. 9.** Modelled daily mean surface concentration of  $\text{NO}_3^-$  and the 12:00 LT wind vector for 12–15 February (episode F), 17–20 March (episode M) and 11–14 April (episode A).

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**Fig. 10.** Mean composition of PM<sub>2.5</sub> components as estimated by the EMEP4UK model for four sites across the UK, averaged for the whole of 2003. The model base run (including all national and international emissions) is compared with the results from a simulation excluding UK emissions. The percentages shown indicate the proportion of total inorganic aerosol at the site that is estimated to due to non-UK emissions. The bars and the percentages account for the contribution of fine primary particulate matter (PPM<sub>2.5</sub>) (shown in black).

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