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Dore, A. J.; Vieno, M.; Tang, Y. S.; Dragosits, U.; Dosio, A.; Weston, K. J.; Sutton, M. A.. 2007 2007 Modelling the atmospheric transport and deposition of sulphur and nitrogen over the United Kingdom and assessment of the influence of SO2 emissions from international shipping. *Atmospheric Environment*, 41 (11). 2355-2367.

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Modelling the atmospheric transport and deposition of sulphur and nitrogen over the United Kingdom and assessment of the influence of SO$_2$ emissions from international shipping

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Abstract

A statistical Lagrangian atmospheric transport model was used to generate annual maps of deposition of sulphur and oxidised and reduced nitrogen for the United Kingdom at a 5 x 5 km^2 resolution. The model was run using emissions for the year 2002. The model was compared with measurements of gas concentrations (SO$_2$, NO$_x$, HNO$_3$ and NH$_3$) and of wet deposition and aerosol concentrations of SO$_4^{2-}$, NO$_3^-$ and NH$_4^+$ from national monitoring networks. Good correlation was obtained, demonstrating that the model is capable of accurately estimating the mass balance and spatial distribution of sulphur and nitrogen compounds in the atmosphere. A future emissions scenario for the year 2020 was used to test the influence of shipping emissions on sulphur deposition in the United Kingdom. The results show that, if shipping emissions are assumed to increase at a rate of 2.5% per year, their relative contribution to sulphur deposition is expected to increase from 9% to 28% between 2002 and 2020. The model was compared to both a European scale and a global scale chemical transport model and found to give broad agreement with the magnitude and location of sulphur deposition associated with shipping emissions. Enforcement of the MARPOL convention to reduce the sulphur content in marine fuel to 1% was estimated to result in a 6% reduction in total sulphur deposition to the UK for the year 2020. The percentage area of sensitive habitats with exceedance of critical loads for acidity in the UK was predicted to decrease by 1% with the implementation of the MARPOL convention.

Keywords: Atmospheric transport model, Acidification, Shipping, Sulphur, Nitrogen, Ammonia

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1. Introduction

The emission of pollutant gases (SO_2, NO_x and NH_3) from the United Kingdom, from European sources and international shipping leads to deposition of acidifying and eutrophying inputs to sensitive ecosystems. Acidification affects soils and freshwater, particularly in upland areas where annual precipitation is high (NEGTAP, 2001). The acidification of soils is known to result in the loss of sensitive species of vegetation and the leaching of cations from soils (Al and Ca^{++}) which are toxic to fish and other biota at high concentrations. Deposition of both reduced nitrogen (NH_x) and oxidised nitrogen (NO_y) results in eutrophication leading to changes in plant species composition and water quality in semi-natural habitats. In addition to these effects, the pollutants provide a source of secondary particulate matter in the atmosphere, through the formation of ammonium sulphate and ammonium nitrate aerosols. The long-range transport of particulate matter means that deposition exceeding the critical loads for environmental damage may occur, even in regions remote from the sources, such as the Scottish Highlands. Aerosols are of additional concern regarding their adverse impacts on human health (COMEAP, 2001).

The need to protect sensitive ecosystems from environmental damage has led to the signing of international and European agreements on reduction of emissions of pollutant gases. These include the **U**nited **N**ation **E**conomic **C**ommission for **E**urope (**UNECE**) Gothenburg protocol, under the **C**onvention on **L**ong-**R**ange **T**ransboundary **A**ir **P**ollution (**CLRTAP**) and the European Community **N**ational **E**missions **C**eiling **D**irective (**NECD**). These agreements lay down targets for nation states to achieve reductions of emissions of SO_2, NO_x and NH_3 by the year 2010. Major reductions to the emissions of SO_2 and NO_x from land sources have been achieved in recent decades in the UK (Dore *et al.*, 2005) and in Europe, with further reductions forecast over the next 15 years (Grice *et al.*, 2005). However the benefit in these emissions reductions is partially offset by the increasing contribution of emissions from international shipping. Estimations of emissions of SO_2

and NO_x from international shipping suggest annual increases of 2.5% (Endresen *et al.*, 2003) caused by increasing traffic and lack of emissions controls. The sulphur content of diesel oil for road traffic was recently lowered by European Community legislation from 350 ppm to 50 ppm. However in European waters, the average sulphur content of marine heavy fuel oil was recently estimated at 2.7% (27,000 ppm) (EEB, 2004). Recent inventories estimate ship emissions of SO_2 to globally contribute between 3.2 Tg [S]/yr (Endresen *et al.*, 2003) to 4.7 Tg [S]/yr (Corbett and Köhler, 2003). These numbers reflect their importance as well as their high uncertainty. Emissions from international shipping fall outside the area of policy influence of the European Community and the UNECE. However the MARPOL convention was put in place by the UN International Maritime Organisation to restrict emissions of SO_2 and NO_x from international shipping. This included a global sulphur cap of 4.5% for bunker fuel and a 1.5% limit for the Baltic Sea and North Sea.

As well as the introduction of monitoring programmes to measure concentrations of pollutants in air and in precipitation, numerical models have been developed to calculate acidic deposition. Atmospheric Transport Models (**ATM**s) simulate the emission of pollutant gases, their chemical transformation in the atmosphere as well as the diffusion and advection of pollutants, their washout by precipitation and dry deposition to vegetation. These models include the EMEP Eulerian model (Tarrasón *et al.*, 2003) which operates at a 50 km resolution and is used to assess acid and nitrogen deposition on a European scale. UK Lagrangian models include FRAME (the Fine Resolution Atmospheric Multi-pollutants Exchange model; Singles *et al.*, 1998), HARM (the Hull Acid Rain Model; Metcalfe *et al.*, 2001) and TRACK (Lee *et al.*, 2000) which operated on 5, 10 and 10 km grid resolutions, respectively. These models used spatial emissions inventories for SO_2, NO_X and NH_3 and were able to calculate deposition of sulphur (SO_x), oxidised nitrogen (NO_y) and reduced nitrogen (NH_x). Estimates of the exceedance of critical loads for acid deposition and nitrogen deposition can subsequently be derived from the modelled deposition. Studies conducted by Derwent *et al.* (2005) used a global Lagrangian chemical transport model to estimate the

contribution to acid deposition in Europe from international shipping emissions along the Atlantic coast.

In this paper we consider the application of FRAME to investigate the distribution of nitrogen and sulphur deposition in the United Kingdom using emissions data for the year 2002. The accuracy of the model was verified by a detailed comparison with measurements from the UK national monitoring networks for gas and particle concentrations in air and ion concentrations in precipitation. The model was applied to demonstrate the important contribution of shipping emissions to sulphur deposition in the United Kingdom. The effect of introduction of a policy to reduce emissions of SO_2 from international shipping in the North Sea, according to the MARPOL convention, was estimated.

2. Description of the Model

The **F**ine **R**esolution **A**tmospheric **M**ulti-pollutant **E**xchange model (**FRAME**) is a statistical Lagrangian trajectory model that can be used to estimate the spatial distribution of sulphur and nitrogen deposition to the United Kingdom. The model has been used for government policy applications in estimating the influence of abatement of pollutant gases, (such as NH_3, NO_x and SO_2) on the exceedance of environmental critical loads for acidification and eutrophication (Vieno, 2005). Conceptually, FRAME represents a column of air with 33 layers of varying thickness ranging from 1 m at the surface and increasing to 100 m thickness at heights above 100 m (Singles *et al.*, 1998). The model domain has a 5 x 5 km^2 horizontal resolution, which includes the United Kingdom and the Republic of Ireland.

The output from FRAME-Europe, a similar model with a 150 x 150 km^2 grid resolution and a domain covering Europe, was used to initialise the FRAME model trajectories. FRAME-Europe uses 50 x 50 km emissions from the EMEP inventory (http://webdab.emep.int/) re-gridded onto the 150 km grid. The directionally-dependent European scale modelled concentrations for all chemical

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variables in a column of air were input to the trajectories at the edge of the FRAME grid. Trajectories were advected across the domain, with different starting angles at a 1° resolution, using directionally dependent wind speed and wind frequency roses generated from a ten year radiosonde data set (Dore *et. al*, 2006). Gridded emissions of SO_2, NO_x and NH_3 were taken from the national atmospheric emissions inventory (www.naei.org.uk, Dore *et al.*, 2005). Vertical diffusion is treated using k-theory eddy diffusivity and solved with a Finite Volume Method (Vieno, 2005). For oxidised nitrogen, a suite of gas phase reactions is considered and both dry phase and aqueous phase oxidation of SO_2 are represented. Wet deposition is calculated with scavenging coefficients and a constant drizzle approach, using precipitation rates calculated from a climatological map of average annual precipitation for the British Isles. An enhanced washout rate is assumed over hill areas due to the scavenging of cloud droplets by the seeder-feeder effect. The washout rate for the orographic component of rainfall is assumed to be twice that calculated for the non-orographic component (Dore *et al.*, 1992). The model incorporates the directional dependence of orographic rainfall described by Fournier *et al.* (2005). Dry deposition of SO_2, NO_2 and NH_3 is calculated individually for five different land cover categories (arable, forest, moor-land, grassland and urban) using a canopy resistance model (Singles *et al.*, 1998). Other species are assigned constant values of deposition velocity.

The FRAME model code is written in High Performance FORTRAN (Fournier *et al.*, 2003) and executed in parallel on a Linux Beowulf cluster. The model has been applied to source-receptor calculations where large numbers of simulations are required. One such example is the estimation of the contribution to nitrogen and sulphur deposition from each of the 75 counties of the UK and from 20 individual major point sources (Oxley *et al.*, 2003).

3. Maps of deposition of sulphur and nitrogen

The most significant data generated from FRAME are maps of dry and wet deposition of sulphur and nitrogen for the United Kingdom, which are used to calculate the exceedance of critical loads for acid deposition and nutrient nitrogen deposition. In previous studies (Fournier *et al.*, 2003, 2005) FRAME has been developed as a model to estimate national scale ammonia concentrations and produce maps of dry and wet deposition of reduced nitrogen. In this study, the model has been developed as multi-pollutant tool. Maps of grid-averaged deposition (including all land cover types) for SO_x, NO_y and NH_x for the year 2002 are illustrated in Figures 1(a) to (f). Figure 1(a) illustrates dry deposition of SO_x, which is principally due to SO_2 gas. Deposition is highest in the industrial regions of northern England and along the east coast where major power stations are sited. Areas of high sulphur deposition also occur in the coastal regions of south and south-east England due to emissions from international shipping in the English Channel. The wet deposition of SO_x (Figure 1(d)) shows a pattern less correlated to the sources, which is a product both of the local scale washout of the soluble gases SO_2 and H_2SO_4 and of the longer-range transport of secondary sulphate aerosol. In Figure 1(b), dry deposition of NO_y is shown to be closely correlated to the major urban areas of Greater London, Birmingham and Manchester and the interconnecting motorways. As the primary pollutants of oxidised nitrogen, NO and NO_2 are non-soluble, wet deposition of NO_y is associated with long range transport of secondary nitrate aerosol and is highest not in the source areas, but in areas of heavy precipitation, particularly the upland areas of Wales and the Pennines in central northern England (Figure 1(e)). Dry deposition of NH_x is illustrated in Figure 1(c). As ammonia is emitted close to ground level and rapidly deposited to vegetation, the map is closely correlated to the emissions. Local variability is evident due to the rapidly varying nature of ammonia emissions over short distances. However, the source areas are evident, resulting in high deposition in western England due to cattle farming and areas of high deposition in the pig and poultry farming regions of eastern England. Significant non-agricultural emissions of NH_3 (Sutton *et al.*, 2000) also

contribute to high deposition of NH_x in major urban areas. Wet deposition of NH_x (Figure 1(f)) is primarily associated with the long-range transport of ammonium aerosol and is therefore highest in hill areas with heavy annual precipitation.

Other methods for generating maps of sulphur and nitrogen deposition for the United Kingdom include application of the EMEP European scale Lagrangian model (Tarrasón *et al.*, 2003) and the **C**oncentration **B**ased **E**stimated **D**eposition technique (**CBED**, Smith *et al.*, 2000; Smith and Fowler, 2001). The latter is a methodology based upon interpolation of wet deposition and gas concentrations measured at sites in the national monitoring networks. CBED dry and wet deposition forms the official UK data set for estimation of exceedance of critical loads for acid deposition and nitrogen deposition. A detailed inter-comparison between these different methodologies will be undertaken in a future study. The CBED and FRAME wet deposition maps generally show similar spatial patterns. In both calculations the wet deposition is driven by a measurement based map of annual precipitation for the UK and includes parameterisation of the orographic enhancement of wet deposition. According to the seeder-feeder effect (Dore *et al.*, 1992) orographic precipitation contains a contribution from the washout of polluted boundary layer hill cloud which has been found by measurements (Beswick *et al.*, 2003) to contain significantly higher concentrations of pollutant ions than non-orographic rainfall samples. The EMEP data shows a much smaller influence of orography on wet deposition. In this case, precipitation is generated by a meteorological model and orographic enhancement of wet deposition is not specifically represented in the model. Despite spatial differences, the average deposition of species show good agreement between FRAME and EMEP with national deposition budgets agreeing to within 20%. The average wet deposition with CBED is higher than FRAME by 13%, 48% and 27% for SO_x, NO_y and NH_x respectively. A notable difference occurs for the NO_y dry deposition budget (46 Gg N year^{-1} for FRAME and 98 Gg N year^{-1} for CBED). This may in part be attributed to an underestimate of HNO_3 concentrations in FRAME.

However uncertainty also arises in the CBED calculation due to the small number of 12 sites in the national monitoring network used for mapping nitric acid concentrations.

4. Comparison of model with measurements from national monitoring networks

Assessment of the accuracy of FRAME in estimating atmospheric concentrations and deposition rates of gaseous and particulate compounds of nitrogen and sulphur can be made by comparison with measurements. For this purpose, data from the twelve sites in the national nitric acid monitoring network (using monthly sampling from the CEH **DELTA** system, **DE**nuder for **L**ong **T**erm **A**tmospheric sampling: Sutton *et al.*, 2001) of gas phase (HNO_3 and SO_2) and aerosol (SO_4^{2-}, NO_3^-) concentrations were applied. Concentrations of NH_3 gas and NH_4^+ aerosol were taken from the UK national ammonia monitoring network (http://www.cara.ceh.ac.uk/nh3network) comprising over 100 DELTA samplers and passive diffusion samplers. Concentrations of NO_2 were taken from the rural monitoring network using diffusion tubes (http://www.aeat.co.uk/netcen). Wet deposition data were obtained from the secondary acid precipitation monitoring network, comprising fortnightly collections of precipitation from 38 sites with ion concentrations analysed by ion chromatography (NEGTAP, 2001). All monitoring data were averaged over a three-year period (2001-2003) to reduce the influence of inter-annual meteorological anomalies. The measurement data were compared with modelled values for the emissions year 2002. The UK national monitoring networks for air quality and acid deposition are described in Hayman *et al.* (2004)

Figures 2(a)-(c) illustrate the correlation with measurements of modelled SO_2 gas concentrations, SO_4^{2-} aerosol concentrations and SO_4^{2-} wet deposition respectively. An overestimate of SO_2 concentrations in more polluted areas is evident from Figure 2(a). Vieno (2005) reported a significant improvement in model correlation with measurements of SO_2 concentrations following introduction of a plume rise parameterisation to the model. However the current overestimate in SO_2 concentrations may indicate either that SO_2 is oxidised too slowly, or mixed to the surface from high

point sources too quickly in the model. For sulphate aerosol (Figure 2(b)), there is a tendency for the model to underestimate low concentrations and overestimate high concentrations. A satisfactory correlation for wet deposition of sulphate is apparent (R^2=0.80, Figure 2(c)), with a slope of 0.95 and low intercept of 0.09 for the model-measurement linear regression.

Figures 3(a)–(d) illustrate the correlation of the model with measurements of oxidised nitrogen species. A high correlation is evident with measurements of NO_2 gas concentrations and NO_3^- aerosol (R^2=0.87, 0.92; Figures 3(a) and 3(c), respectively). However, the model underestimates wet deposition of nitrate (Figure 3(d)) which may indicate either that the washout coefficient for nitrate aerosol used in the model is underestimated or that the conversion of NO_x gas to nitrate aerosol is proceeding too slowly. The data for measurement of HNO_3 concentrations show greater deviation from modelled concentrations (Figure 3(b)). Although agreement is seen at concentrations of about 0.5 $\mu g\ m^{-3}$, the line of best fit suggests an underestimate in concentrations of HNO_3 by the model at higher concentrations. The two locations where the modelled concentrations are much lower than measurements are sites with agricultural activity which suggests uncertainty in the HNO_3 interaction with NH_3. In FRAME and according to theory, HNO_3 concentrations are rapidly depleted in the presence of NH_3 by the conversion from gas to particulate ammonium nitrate. This process may be subject to significant sub-grid variability due to the highly localised distribution of NH_3 emissions.

Figures 4(a)–(c) illustrate the correlation of the model with measurements of reduced nitrogen species. The correlation of modelled concentrations of NH_3 with measurements (Figure 4(a)) shows considerable scatter. The principal reason for this is the highly localised nature of NH_3 emissions, such that the modelled average concentration from a 5 km model grid cell may differ significantly from that measured at a specific location within the grid cell (Dragosits *et al.*, 2002). The graph shows evidence that, particularly at low concentrations, the model overestimates NH_3 surface concentrations. There is a need for finer scale national modelling of ammonia

concentrations, preferably at a 1 km resolution, in order to perform a more accurate model-measurement comparison. A better correlation is observed between modelled and measured NH_4^+ concentrations (Figure 4(b)) and wet deposition (Figure 4(c)). This is due to the more slowly changing pattern in NH_4^+ aerosol concentrations, which are not expected to vary on a scale smaller than the 5 km model grid resolution. Figure 4(b) shows that the model generally underestimates NH_4^+ aerosol concentrations which may indicate that the rate of production of NH_4^+ aerosol from NH_3 gas is underestimated in the model.

The results of correlation with measurements are illustrated for all species in Table 1. Overall, the correlations give reasonable slopes and high values of the correlation coefficient R^2. This suggests that FRAME is able to accurately represent the spatial distribution of atmospheric gas and particle concentrations and deposition fluxes over the United Kingdom. The verification of the model with measurements suggests that it is an appropriate tool to be used for testing the influence of policies to abate emissions of SO_2, NO_x and NH_3.

5. Policy Application of FRAME to the role of shipping emissions

The contribution of international shipping emissions to air pollution has recently received much attention (Johnson *et al.*, 2000). Whilst land-based emissions in Europe have shown significant downward trends over recent decades, shipping emissions of SO_2 are estimated to be increasing at a rate of 2.5 % per year. Emissions of SO_2 from international shipping in the coastal regions of the British Isles are illustrated in Figure 5(a), gridded at a 50 km resolution (ENTEC, 2002; data available from: www.emep.int). The intensity of emissions is highest in the busy shipping lanes of the English Channel and in the vicinity of major ports. The relative contribution of shipping emissions to sulphur deposition in the United Kingdom was assessed by conducting two model simulations. The first simulation included all sources (UK national emissions, long range transport of pollutants from Europe and shipping emissions). In the second simulation, shipping emissions

were set to zero. The difference in modelled sulphur deposition between the two simulations represents the contribution from shipping emissions alone (Figure 5(b)). The significant contribution to sulphur deposition in the south-east of England and in other coastal regions is apparent. This close-to-source contribution is principally due to the dry deposition of SO_2 gas to vegetation. However, a long range transport component of sulphur deposition due to shipping can also be seen in the form of high deposition in the hill regions of Wales and northern England. This occurs due to the oxidation of SO_2 to sulphate aerosol, which is removed from the atmosphere principally by washout from precipitation and results in high deposition in the high rainfall upland regions. This procedure was applied for both the year 2002 and for a future scenario for the year 2020 using emissions from the UK Air Quality Strategy (Grice *et al.*, 2005). The future scenario included the significant forecast reductions in land-based emissions relative to 2002 (of 72%, 45% and 10% for SO_2, NO_x and NH_3 respectively) as well as the increase in shipping emissions (Table 2). The forecast emissions for the year 2020 also include significant reductions beyond the targets set by the NECD for the UK for the year 2010 (of 45%, 23% and 9% for SO_2, NO_x and NH_3 respectively). Total emissions for the two years are shown in Table 2. The results of the model simulations, illustrated in Table 3, suggest that the relative contribution of shipping emissions to sulphur deposition in the United Kingdom will increase from 9% to 28% over the period 2002-2020, with similar contributions to both wet and dry deposition.

The results obtained with FRAME compare favourably with estimates from the EMEP model (Klein *et. al*, 2005) which suggest that shipping emissions contribute 13 % to sulphur deposition for the year 2002. In the EMEP model, the larger 50 km grid cells result in a greater proportion of the UK land surface area being represented by coastal grid cells, where emissions from the sea occur simultaneously with the process of deposition to land surface. As FRAME is a national scale model, the finer 5 km resolution of the grid cells permits a better distinction between land and sea. A global three-dimensional Lagrangian chemistry-transport model (STOCHEM) run at a 5° x 5° resolution

was applied by Derwent *et al.* (2005) to assess the regional acid deposition along the Atlantic seaboard of Europe. Total sulphur deposition was found to exceed 2 kg S ha^{-1} in the south-west of the UK. The results with FRAME (figure 5(b)) also exhibit significant areas in the south and west of the country with the contribution to total sulphur deposition exceeding 2 kg S ha^{-1}. However, the two models used very different horizontal grid spacing and a direct comparison between results would require regridding of FRAME data onto the STOCHMEM grid.

The significance of shipping emissions in contributing to sulphur deposition over land lends strong support to the need for international legislation to constrain emissions from shipping, such as the **MARPOL** convention on **MAR**ine **POL**lution. Recent legislation involves the introduction of a 1 % sulphur limit on marine fuels used by all sea-going vessels in the North Sea and the English Channel from the year 2010, leading to a 33% reduction in emissions of SO_2. An additional model simulation was undertaken for the year 2020 with SO_2 emissions from shipping reduced in accordance with the MARPOL convention. The abatement measure was effective in reducing total sulphur deposition to the UK from 86 Mg to 78 Mg and reducing the relative contribution of shipping emissions to UK sulphur deposition from 28% to 20%. The changes to exceedance of critical loads for acid deposition to the environment between 2002 and 2020 have been estimated by Hall *et al.* (2006) using the FRAME modelled deposition data. As part of the UK Air Quality Strategy, a number of additional emissions abatement scenarios were considered individually, including the implementation of the MARPOL convention for international shipping emissions, emissions of NO_x from large and small combustion plants and a variety of measures to reduce emissions of NO_x from road vehicles (Stedman *et al.*, 2006). The percentage area of ecosystems in the UK subject to acid deposition above their critical loads was calculated to decrease from 54.8% to 39.4% during the period 2002 to 2020. The additional implementation of the MARPOL convention was found to further reduce the area of ecosystems with exceedance of acidity critical loads to 38.5%, an improvement of similar magnitude to a scenario which combined the introduction of low

emission vehicles with reduced emissions of NO_x from small combustion plants. However, abatement of NOx emissions had the additional benefit of reducing exceedance of critical loads for eutrophication. These results suggest that targeting shipping emissions may be an effective way of protecting the environment from acid deposition.

Whilst both SO_2 and NO_x emissions have shown strong downward trends in emissions in recent decades, reductions in NH_3 emissions have been smaller, and driven primarily by the influence of market forces on national agricultural activities. The growing relative importance of reduced nitrogen to total acidic and total nitrogen deposition indicates that future strategies to tackle acidification and eutrophication will need to include measures to abate emissions of ammonia.

Conclusion

The Fine Resolution Atmospheric Multi-pollutant Exchange model (FRAME) was applied to estimate the deposition of sulphur and oxidised and reduced nitrogen to the United Kingdom using emissions data for the year 2002. The model showed satisfactory agreement with measurements of gas and aerosol concentrations and wet deposition of nitrogen and sulphur compounds from the national monitoring networks. The results suggest that the model is an appropriate tool for testing the effects of policies to abate emissions of SO_2, NO_x and NH_3.

The growing importance of SO_2 emissions from international shipping contrasts with the strong downward trend in land based SO_2 emissions. In the absence of emissions controls, international shipping is set to become a major source of sulphur deposition, particularly in south England. The model simulations for a recent emissions year and for a future scenario, assuming a 2.5% increase in emissions from international shipping per year, suggest that the contribution of shipping emissions to total sulphur deposition in the United Kingdom will increase from 9% to 28% between 2002 and 2020. The modelled relative contribution of shipping emissions to total sulphur deposition in the UK for 2002 was also in broad agreement to calculations made with the European

scale EMEP model (13%). The model reproduced the maximum deposition of sulphur from shipping emissions of up to 2 kg S Ha^{-1} in southern and western England estimated with the STOCHEM global scale model.

The influence of enforcing the MARPOL convention to restrict the sulphur content of bunker fuel used by international shipping in the North Sea from 1.5% to 1% was tested with the model. This was found to result in a 6% reduction in total sulphur deposition to the United Kingdom for the year 2020 and a reduction in the percentage of land with exceedance of critical loads for acid deposition from 39.4% to 38.5%. This improvement to acid deposition was of similar magnitude to results obtained with other abatement strategies targeted at emissions of NO$_x$ from vehicles and combustion plants, suggesting that controlling emissions from international shipping may be an efficient way to reduce the influence of acid deposition.

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Table 1: Parameters for the linear regression $y_{(modelled)} = m * x_{(measured)} + c$
R^2 is the correlation coefficient. Concentrations are in $\mu g\ m^{-3}$ and wet deposition in kg N or S ha^{-1}

	m	c	R^2
SO_2 concentration	1.22	+0.11	0.88
SO_4^{--} concentration	1.25	-0.20	0.83
NO_2 concentration	0.96	-0.25	0.87
NO_3^{--} concentration	0.97	-0.21	0.92
NH_3 concentration	0.96	+0.75	0.58
NH_4^+ concentration	0.60	-0.11	0.85
HNO_3 concentration	0.57	+0.13	0.81
SO_4^{--} wet deposition	0.95	-0.09	0.80
NO_3^- wet deposition	0.75	-0.02	0.77
NH_4^+ wet deposition	0.94	-0.11	0.77

Table 2: Total emissions of SO_2, NO_x and NH_3 from the UK and international shipping for the year 2002 and for the year 2020 according to the Air Quality Strategy projections

	SO_2 land emissions (kT S)	SO_2 shipping emissions (kT S)	NO_x land emissions (kT N)	NH_3 land emissions (kT N)
2002	501	97	481	248
2020	162	151	272	224

Table 3: The modelled total sulphur deposition budget to the United Kingdom and contribution originating from emissions of SO_2 from international shipping. Three scenarios are considered: (i) for the year 2002 , (ii) for the year 2020 assuming a Business As Usual scenario (BAU) and (iii) assuming application of the MARPOL convention for the year 2020.

SO_x deposition	2002	2020 BAU	2020 MARPOL
Dry deposition (Mg S)	55	27	25
Wet deposition (Mg S)	104	59	55
Total deposition (Mg S)	159	86	81
Deposition from shipping emissions (Mg S)	15	24	19
% contribution to total deposition from shipping emissions	9.3	28	23

Figure 1(a): 2002 grid average dry deposition of SO_x (kg S ha^{-1})
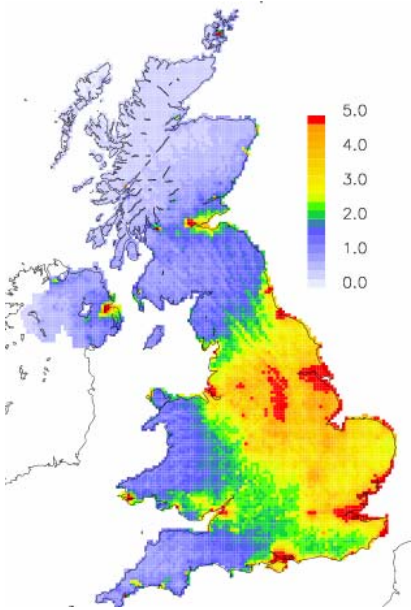
Figure 1(b): 2002 grid average dry deposition of NO_y (kg N ha^{-1})
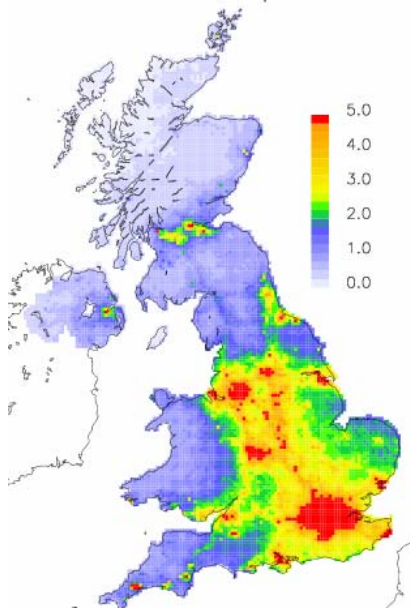
Figure 1(c): 2002 grid average dry deposition of NH_x (kg N ha^{-1})
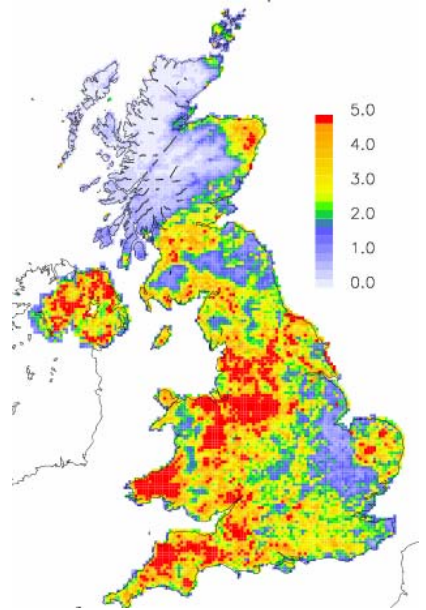
Figure 1(d): 2002 wet deposition of SO_x (kg S ha^{-1})
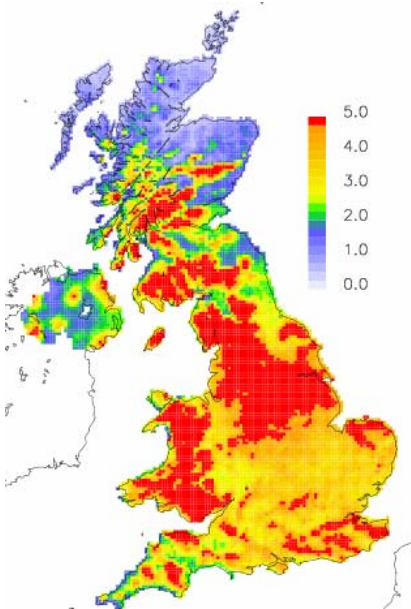
Figure 1(e): 2002 wet deposition of NO_y (kg N ha^{-1})
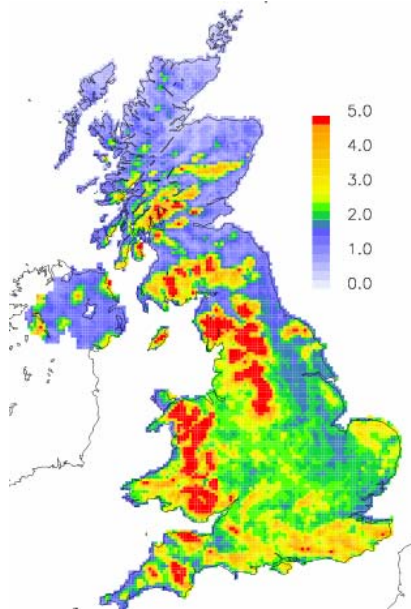
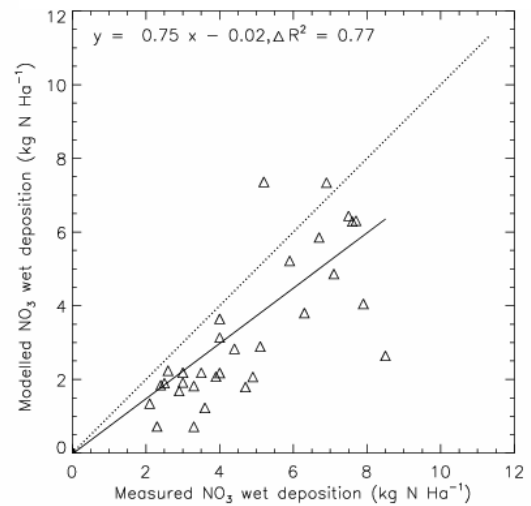**Figure 1(f):** 2002 wet deposition of NH_x (kg N ha^{-1})
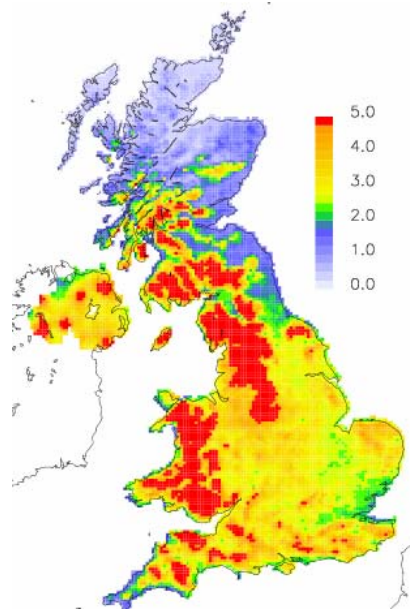
Figure 2(a) Correlation of modelled concentrations of SO_2 with measurements from the national monitoring network
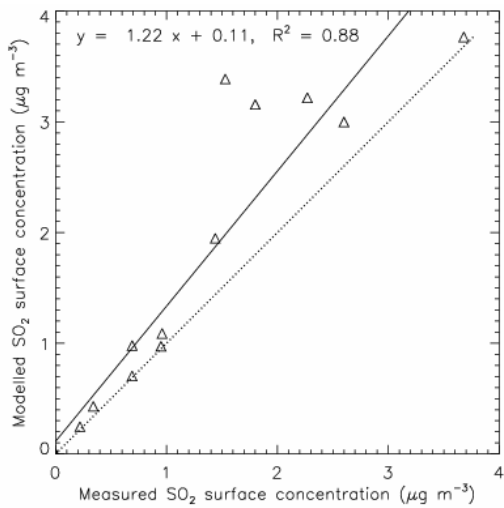
Figure 2(b) Correlation of modelled concentrations of SO_4^{2-} aerosol with measurements from the national monitoring network
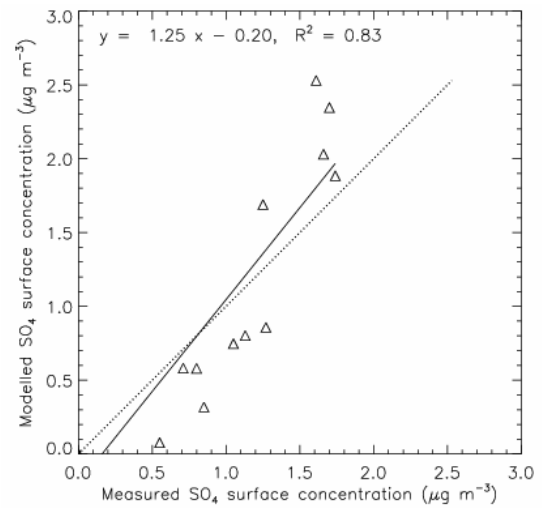
Figure 2(c) Correlation of modelled SO_4^{2-} wet deposition with measurements from the national monitoring network
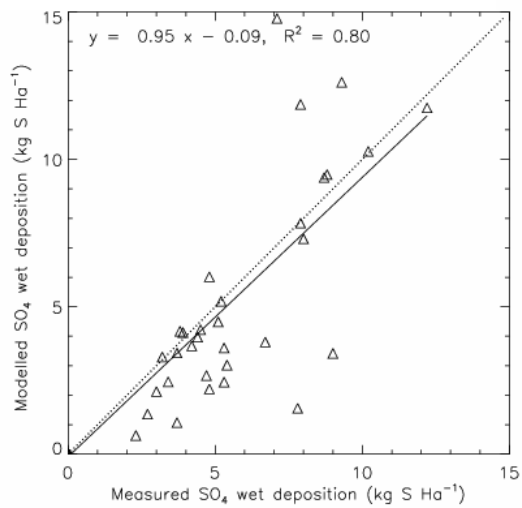
Figure 3(a) Correlation of modelled concentrations of NO_2 with measurements from the national monitoring network
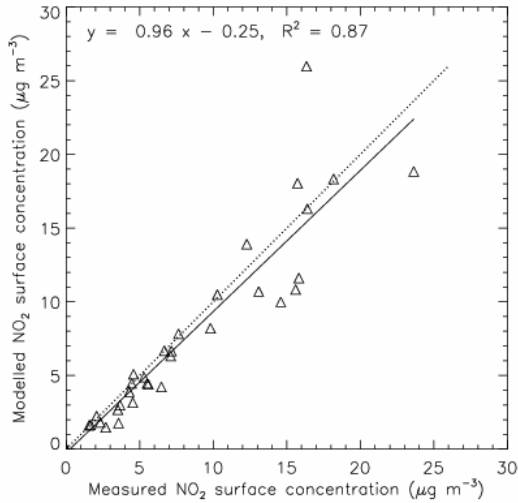
Figure 3(b) Correlation of modelled concentrations of HNO_3 with measurements from the national monitoring network
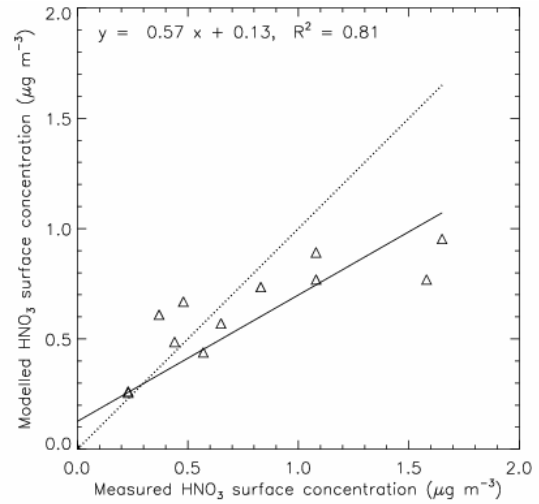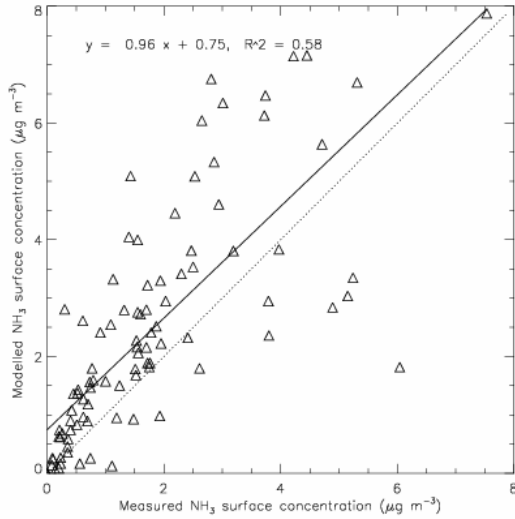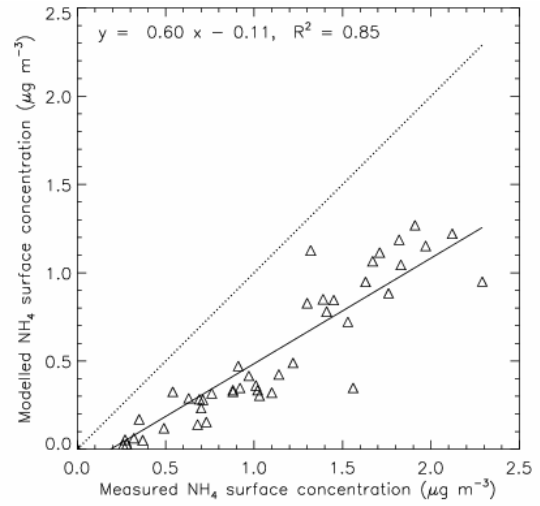

Figure 3(c) Correlation of modelled concentrations of NO_3^- aerosol with measurements from the national monitoring network
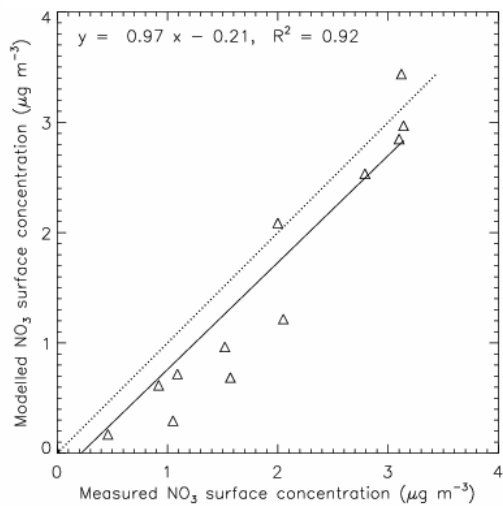
Figure 3(d) Correlation of modelled NO_3^- wet deposition with measurements from the national monitoring network
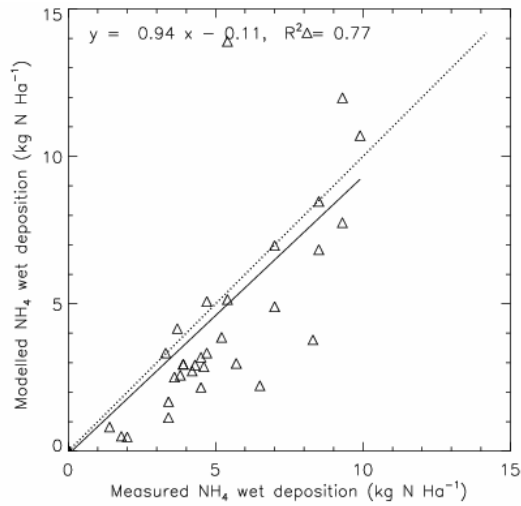
Figure 4(a) Correlation of modelled concentrations of NH$_3$ with measurements from the national monitoring network

Figure 4(b) Correlation of modelled concentrations of NH$_4^+$ aerosol with measurements from the national monitoring network
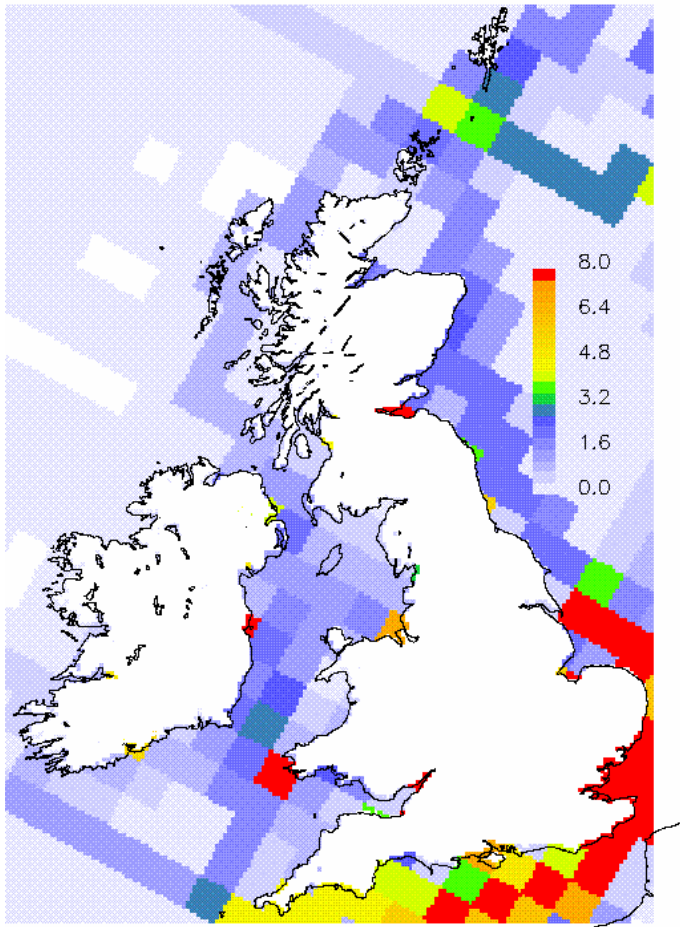

Figure 4(c) Correlation of modelled NH$_4^+$ wet deposition with measurements from the national monitoring network

Figure 5(a) Emissions of SO$_2$ from international shipping for the year 2002 within the FRAME domain, gridded at a 50 km resolution (kg S ha^{-1} yr^{-1})

Figure 5(b) FRAME modelled deposition of SO$_x$ from international shipping emissions for the year 2002 (kg S ha^{-1} yr^{-1})