MODELLING THE CONTRIBUTION OF SO_2 AND NO_X EMISSIONS FROM INTERNATIONAL SHIPPING TO SULPHUR AND OXIDISED NITROGEN DEPOSITION IN THE UNITED KINGDOM

Anthony J. Dore[1], Maciej Kryza[2], Massimo Vieno[3] and Mark A. Sutton[1]

[1] Centre for Ecology and Hydrology, Penicuik, Midlothian EH26 9HF, UK
[2] Department of Meteorology, University of Wrocław,
ul. Kosiby 8, 51-670 Wroclaw, Poland
[3] Institute of Atmospheric and Environmental Science, Crew Building,
The University of Edinburgh, King's Buildings, Edinburgh EH9 3JN, UK

Abstract: A statistical Lagrangian atmospheric transport model was used to generate annual maps of deposition of sulphur and nitrogen for the United Kingdom at a 5 x 5 km^2 resolution for the year 2005 and to assess the contribution attributed to emissions of SO_2 and NO_x from international shipping. A future emissions scenario for the year 2020 was used to investigate changes in the relative contribution of shipping emissions. The results show that, if shipping emissions are assumed to increase at a rate of 2.5% per year, their relative contribution to total sulphur and oxidised nitrogen deposition are expected to increase from 15% and 12% respectively to 37% and 28% between 2005 and 2020. Enforcement of the International Maritime Organisation (IMO) agreement to reduce the sulphur content in marine fuel to 0.5% was estimated to result in a 30% reduction in total sulphur deposition to the UK for the year 2020 compared to a business as usual scenario, with the result that the relative contribution from shipping to sulphur deposition in the UK would be reduced to 9% of the total

Key words: *Atmospheric transport model, Acidification, Shipping, Sulphur, Nitrogen.*

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 affecting soils and freshwater, particularly in upland areas where annual precipitation is high. 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. 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 agreements lay down targets for nation states to achieve reductions of emissions of SO_2, NO_x and NH_3. 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. In European waters, the average sulphur content of marine heavy fuel oil was recently estimated at 2.7% (EEB, 2004). Estimates of emissions of SO_2 and NO_x from international shipping remain subject to high uncertainty, whilst mapping the emissions presents additional problems not associated with land sources. The Marine Environment Protection Committee (MEPC) of the International Maritime Organisation (IMO) recently decided that the sulphur content of all marine fuels will be capped at 0.5 per cent worldwide from 2020.

In this paper we consider the application of a Lagrangian atmospheric transport model, FRAME, to investigate the distribution of sulphur and oxidised nitrogen deposition in the United Kingdom. Emissions data for the year 2005 and projections for 2020 were used to assess the changing contribution of shipping emissions to sulphur and nitrogen deposition in the United Kingdom both with and without controls on sulphur content in fuel.

2. OVERVIEW OF THE FRAME MODEL

The main features of the FRAME model can be summarised as:

- 5 x 5 km^2 resolution over the British Isles (incorporating the Republic of Ireland); grid dimensions: 244 x 172.
- Input gas and aerosol concentrations at the edge of the model domain are calculated with FRAME-Europe, using European emissions and run on the EMEP 50 km scale grid.
- Air column divided into 33 layers moving along straight-line trajectories in a Lagrangian framework with a 1^o angular resolution. The air column advection speed and frequency for a given wind direction is statistically derived from radio-sonde measurements (Dore *et al.*, 2006). Variable layer thickness from 1 m at the surface to 100 m at the top of the mixing layer.
- Emissions are gridded separately by SNAP sector for SO_2 and NO_x and by agricultural sector for NH_3 and injected into vertical model layers which depend on the sector.
- Vertical diffusion in the air column is calculated using K-theory eddy diffusivity and solved with the Finite Volume Method.
- Wet deposition is calculated using a diurnally varying scavenging coefficient depending on mixing layer depth and a 'constant drizzle' approximation driven by an annual rainfall map. A precipitation model is used to calculate wind-direction-dependent orographic enhancement of wet deposition (Fournier *et al.*, 2005)

- Five land classes: forest, moorland, grassland, arable, urban & water are considered. A vegetation specific canopy resistance parameterisation is employed to calculate dry deposition of SO_2, NO_x and NH_3.
- The model chemistry includes gas phase and aqueous phase reactions of oxidised sulphur and oxidised nitrogen and conversion of NH_3 to ammonium sulphate and ammonium nitrate aerosol.

3. MODEL RESULTS AND COMPARISON WITH MEASUREMENTS

The data generated from FRAME include maps of dry and wet deposition of sulphur and nitrogen for the United Kingdom, which have been used to calculate the exceedance of critical loads for acid deposition and nutrient nitrogen deposition. The relatively high speed of simulation makes the model suitable for application to studies involving large numbers of simulations such as integrated assessment modelling (Oxley *et al.* 2003) and uncertainty studies as well as addressing policy questions on future emissions controls.

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).

The results of the correlation with measurements are summarised in Table 1. Generally the model is able to obtain a good correlation with annual average measurements of concentrations of gaseous and particulate compounds in air and with wet deposition. There is some over-estimate of concentrations of SO_2 and sulphate aerosol by the model. Nitric acid concentrations are underestimated by the model, a feature commonly observed in chemical transport models. There is also an underestimate of ammonium aerosol, either due to an underestimate in the component imported from the large scale European simulation or a slow formation rate from ammonia gas. The scatter is highest in the correlation with ammonia concentrations due to the highly localised variability in emissions which is not captured with the model 5 km grid resolution.

Table 1. Parameters for the linear regression $y_{(modelled)} = m * x_{(measured)} + c$
R^2 is the correlation coefficient. Concentrations are in μgm^{-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

4. APPLICATION OF THE MODEL TO ASSESSMENT OF 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 and NO_x are estimated to be increasing at a rate of 2.5% per year. Emissions of SO_2 from international shipping in the region of the FRAME-UK and FRAME-Europe model domains are illustrated in Figures 1(a) and 1(b) respectively, 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 and oxidised nitrogen deposition in the United Kingdom

was assessed by conducting two sets of model simulations. The first set of simulation included all sources (UK national emissions, long range transport of pollutants from Europe and shipping emissions). In the second set, shipping emissions of SO_2 and NO_x were set to zero in both the regional scale (5km resolution) and the European scale (50 km resolution) simulations. The difference in modelled sulphur and nitrogen deposition between the two simulations represents the contribution from shipping emissions alone. The significant contribution to sulphur dry deposition in the south-east of England (Fig. 2(a)) 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 wet 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 (Fig. 2(b)). This procedure was applied for both the year 2005 and for a future scenario for the year 2020 using emissions from the UK Air Quality Strategy (Grice *et al.*, 2005). Emissions for the UK for 2005 were 362 Gg S-SO_2 and 499 Gg N-NO_x. Shipping emissions from the FRAME UK model domain (including a large part of the North Sea and English Channel were 94 Gg S-SO_2 and 89 Gg N-NO_x. The future scenario included the significant forecast reductions in land-based emissions relative to 2005 (of 60%, 34% and 10% for SO_2, NO_x and NH_3 respectively) as well as the increase in shipping emissions. The results of the model simulations, illustrated in Table 2, suggest that, in the absence of emissions controls on international shipping, the relative contribution of shipping emissions to sulphur and oxidised nitrogen deposition in the United Kingdom will increase from 15% to 37% and 12% to 28% respectively over the period 2005-2020 (although total deposition will decrease due to the reductions in land-based emissions).

The results obtained with FRAME compare favourably with estimates from the EMEP model (Klein *et. al*, 2005) which suggest that shipping emissions contribute 19% and 18% respectively to sulphur and oxidised nitrogen deposition for the year 2005. 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. It may therefore be expected that the EMEP model would obtain higher values for the shipping contribution to land deposition.

The significance of shipping emissions in contributing to oxidised nitrogen and sulphur deposition over land lends strong support to the need for international legislation to constrain emissions from shipping. The IMO has recently made an agreement to reduce the sulphur content in marine fuel to 0.5% by 2020, a significant reduction from currently much higher levels of 2.7% on average. An additional model scenario was run with emissions of SO_2 assumed to fall corresponding to the reduction in sulphur content. The result of this policy gives a clear benefit with the total UK sulphur deposition budget in 2020 falling from 122 to 85 Gg S. This can be expected to result in a significant decrease in exceedance of critical loads for acid deposition to ecosystems. It is important to note however that increases in shipping emissions based on the assumed growth of traffic of 2.5% per year between 2005 and 2020 are based on global estimates. The real influence of shipping emissions to the UK will depend on future changes of traffic through the English channel.

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.

Table 2. The modelled total sulphur and oxidised nitrogen deposition budgets to the United Kingdom and contribution originating from international shipping emissions. Three scenarios are considered: (i) for the year 2005, (ii) for the year 2020 assuming a Business As Usual scenario (BAU) and (iii) assuming application of the IMO agreement for the year 2020.

	2005		2020 BAU		2020 IMO	
	SO_x	NO_y	SO_x	NO_y	SO_x	NO_y
Dry deposition (Gg S/N)	60	71	38	44	26	44
Wet deposition (Gg S/N)	125	104	84	71	59	71
Total deposition (Gg S/N)	185	175	122	115	85	115
Deposition from shipping emissions (Gg S/N)	28	21	45	32	8	32
% contribution to total deposition from shipping emissions	15	12	37	28	9	28

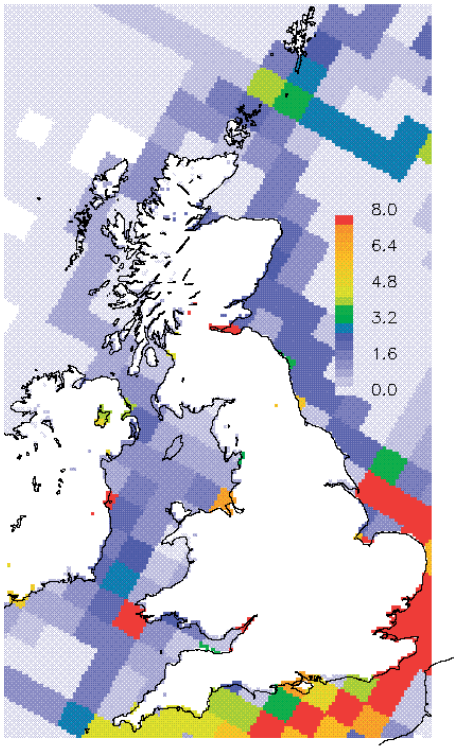

Figure 1(a). Emissions of SO_2 from international shipping in the FRAME-UK domain (kg S Ha^{-1})

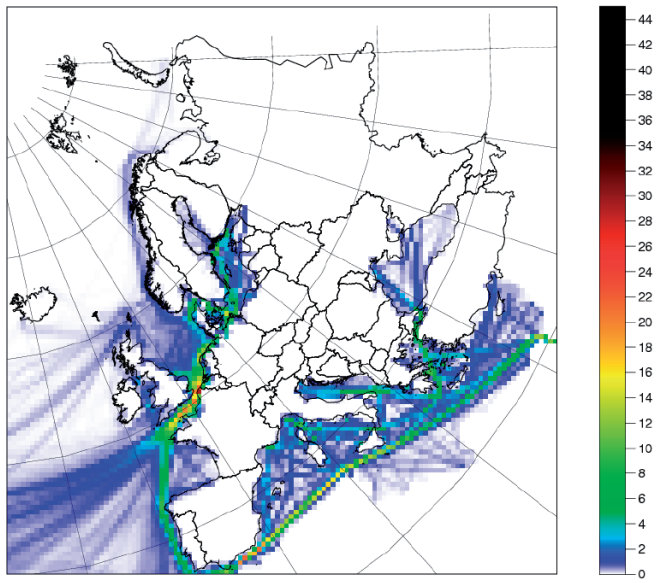

Figure 1(b). Emissions of SO_2 from international shipping in the FRAME-Europe domain (kg S Ha^{-1})

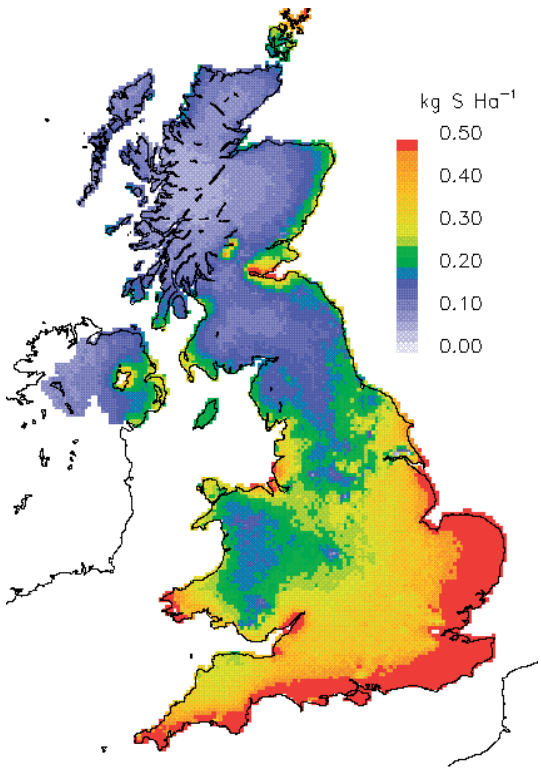

Figure 2(a). Dry deposition of SO_x due to emissions from international shipping (kg S Ha^{-1})

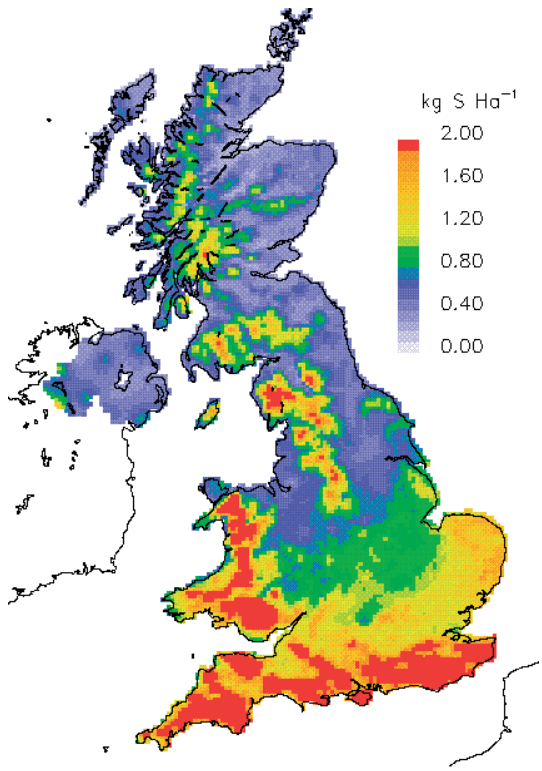

Figure 2(b). Wet deposition of SO_x due to emissions from international shipping (kg S Ha^{-1})

5. CONCLUSION

A Lagranian atmospheric transport model has been used to estimate the deposition of sulphur and nitrogen to the UK. The model showed that emissions of SO_2 and NO_x from international shipping make contributions of 15% and 12% to total deposition of sulphur and oxidised nitrogen in the UK respectively. With traffic from international shipping expected to continue to increase over the next decade, emissions controls on shipping are shown to be necessary in order to continue recent progress in Europe to reduce acid deposition and nitrogen deposition. Considerable uncertainty in both the magnitude and spatial distribution of shipping emissions remains. Further work is required to address these uncertainties and also to provide accurate future estimates of shipping emissions and more finely resolved spatial maps.

REFERENCES

Dore, A.J., Vieno, M., Fournier, N., Weston, K.J. and Sutton, M.A., 2006: Development of a new wind rose for the British Isles using radiosonde data and application to an atmospheric transport model. *Q.J.Roy.Met.Soc.* (in press)

Dore, C.J., J D Watterson, T P Murrells, N R Passant, M M Hobson, S L Baggott, G Thistlethwaite, J W L Goodwin, K R King, M Adams, C Walker, M K Downes, P J Coleman, R A Stewart, A Wagner, J Sturman, C Conolly, H Lawrence and P R Cumine, 2005: UK Emissions of Air Pollutants 1970 to 2003. AEA Technology http://www.airquality.co.uk/archive/reports/cat07/.

EEB, 2004: Air Pollution from ships. Briefing document, the European Environment Bureau.

ENTEC, 2002: Quantification of emissions from ships associated with ship movements between ports in the European Community. Study for the European Commission (www.europa.eu.int/comm/environment/air/background.htm#transport)

Endresen, Ø., E. Sørgård, J. K. Sundet, S. B. Dalsøren, I. S. A. Isaksen, T. F. Berglen, and G. Gravir, 2003: Emission from international sea transportation and environmental impact. *J. Geophys. Res.,* **108(D17)**, 4560, DOI:10.1029/2002JD002898.

Fournier, N., Weston, K.J., Dore, A.J. and Sutton, M.A., 2005: Modelling the wet deposition of reduced nitrogen over the British Isles using a Lagrangian multi-layer atmospheric transport model. *Q.J.Roy.Met.Soc.*, **131**, 703-722 .

Grice, S., T. Bush, J. Stedman, K. Vincent, A. Kent, J. Targa, 2005: Baseline Projections of Air Quality in the UK for the 2005 Review of the Air Quality Strategy. *AEA Technology*, AEAT/ENV/R/1936 Issue 1, April 2005.

Hayman, G., K.J. Vincent, H. Lawrence, M. Smith, M. Davies, M.A. Sutton, Y.S. Tang, U. Dragosits, L. Love. and D. Fowler, 2004: Management and Operation of the UK Acid Deposition Monitoring Network: Data Summary for 2003. Department for the Environment, Food and Rural Affairs, Report number AEAT/ENV/R/1818.

Johnson, J.E., Tarrasón, L. and Bartnicki, J., 2000: Effects of international shipping on European pollution levels. *EMEP/MSC-W Note* **5/00**. EMEP, Meteorological Synthesizing Centre – West, Norwegian Meteorological Institute, Oslo, Norway (www.emep.int)

Klein, H., P. Wind and M. van Loon, 2005: Transboundary air pollution by main pollutants (S, N, O_3) and PM: The United Kingdom. *MSC-W Data Note* **1/2005**, Norwegian Meteorological Institute.

Oxley, T. ApSimon, H., Dore, A.J., Sutton, M.A. Hall, J., Heywood, E., Gonzales del Campo, T. and Warren, R., 2003: The UK Integrated Assessment Model, UKIAM: A National Scale Approach to the analysis of strategies for abatement of atmospheric pollutants under the convention on long-range transboundary air pollution. *Integrated Assessment*, **4**, 236-249.

Sutton, M.A., Tang, Y.S., Miners, B. and Fowler, D., 2001: A new diffusion denuder system for long-term, regional monitoring of atmospheric ammonia and ammonium. *Water, Air and Soil Pollution: Focus*, **1**, 145-156.