



The importance of source configuration in quantifying footprints of regional atmospheric sulphur deposition

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ABSTRACT

An atmospheric transport-chemistry model is applied to investigate the effects of source configuration in simulating regional sulphur deposition footprints from elevated point sources. Dry and wet depositions of sulphur are calculated for each of the 69 largest point sources in the UK. Deposition contributions for each point source are calculated for 2003, as well as for a 2010 emissions scenario. The 2010 emissions scenario has been chosen to simulate the Gothenburg protocol emission scenario. Point source location is found to be a major driver of the dry/wet deposition ratio for each deposition footprint, with increased precipitation scavenging of SO_x in hill areas resulting in a larger fraction of the emitted sulphur being deposited within the UK for sources located near these areas. This reduces exported transboundary pollution, but, associated with the occurrence of sensitive soils in hill areas, increases the domestic threat of soil acidification. The simulation of plume rise using individual stack parameters for each point source demonstrates a high sensitivity of SO₂ surface concentration to effective source height. This emphasises the importance of using site-specific information for each major stack, which is rarely included in regional atmospheric pollution models, due to the difficulty in obtaining the required input data. The simulations quantify how the fraction of emitted SO_x exported from the UK increases with source magnitude, effective source height and easterly location. The modelled reduction in SO_x emissions, between 2003 and 2010 resulted in a smaller fraction being exported, with the result that the reductions in SO_x deposition to the UK are less than proportionate to the emission reduction. This non-linearity is associated with a relatively larger fraction of the SO₂ being converted to sulphate aerosol for the 2010 scenario, in the presence of ammonia. The effect results in less-than-proportional UK benefits of reducing in SO₂ emissions, together with greater-than-proportional benefits in reducing export of UK SO₂ emissions.

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1. Introduction

Computer modelling techniques are now routinely applied to estimate concentrations and deposition of air pollutants in the natural environment. Numerical models have a number of advantages over measurement-based techniques: they provide good spatial coverage over the domain of interest; they can be applied to estimate environmental impacts for future emissions scenarios; they can be used to attribute pollutant emissions sources to concentrations and deposition; they are important for integrated assessment modelling of cost-effective techniques to achieve emissions reductions leading to protection of the environment; and their application often represents a fast and low-cost approach. Numerical models have been applied at

different spatial resolutions to estimate acid deposition over Europe with a 50 km resolution Eulerian model (e.g., Simpson et al., 2003; <http://www.emep.int>) and over the UK with statistical Lagrangian models (e.g., Metcalfe et al., 2001; Fournier et al., 2005; Dore et al., 2007a).

Under the UK Conservation Regulations (known as the Habitats Regulations), which represent the UK implementation of the Habitats Directive, existing and new consents given to industry must be reviewed by competent bodies to demonstrate that installations have no adverse effect on the ecosystems of "Natura 2000" sites established under the Directive. For any consents which are likely to have a significant or adverse effect on a European site (e.g., Special Area of Conservation, SAC or Special Protection Area, SPA), either individually or in combination with others, an appropriate assessment of the implications for the site should be carried out.

Atmospheric modelling tools are needed that are sufficiently robust for such assessments, but sufficiently simple to be applied many times for different point sources. To implement such activity in the UK, we have developed a multi-layer statistical Lagrangian model

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of the British Isles (FRAME, Singles et al., 1998). In principle, it is has long been known that source configuration affects the extent of atmospheric transport, with high stack emissions and plume rise reducing ground level concentrations (Seinfeld and Pandis, 1997). Representation of plume rise is well developed in local dispersion models. However, long-range transboundary air pollution models applied at national and continental scales do not often specify these factors explicitly for the major point sources (e.g., EMEP model, Simpson et al., 2003). For example, treatment of these factors is difficult to achieve where vertical resolution is not specified in detail (e.g., HARM model – Metcalfe et al., 2001). However, a more recent version of the HARM model (Metcalfe et al., 2005) has been developed to include a more detailed vertical structure allowing pollutants to be injected at specific heights. In models with explicit representation of vertical mixing, a lack of site-based data for point sources often prevents the implementation of a detailed scheme related to source height and plume rise.

In this study for the UK, stack parameters for the main sources of SO₂ emission were obtained in cooperation with each of the devolved authorities for England and Wales, Scotland and Northern Ireland (Dore et al., 2007b), including stack height, exit temperature and exit velocity, specified for each of the 69 largest SO_x sources in the UK. The SO_x point sources included in this study account for ~92% of the total UK SO_x point source emissions and ~80% of all SO_x UK sources. The number of sources was selected to be optimal in restricting the number of footprint calculations undertaken whilst including most big sources of sulphur emissions. This has enabled us to investigate the role of source height configuration in affecting the fate of SO_x emissions on a national scale. Using this approach, we here simulate deposition “footprints” associated with SO_x emissions for the year 2003 and future estimates for 2010. We then use these simulations to examine the role of source configuration on patterns of UK sulphur deposition and export. The 2010 scenario was calculated based on future predictions of energy consumption and taking into account use of low sulphur fuel (including use of gas in place of coal) as well as large emissions reductions for specified power stations due to retrofit of flue gas desulphurization (FGD).

Local dispersion models have been used to look at the foot prints of individual point sources in past studies. However, a mesoscale model, which includes atmospheric chemical reactions, is needed in order to consider the consequences for wet deposition due to the role of long range transport. Moreover, mesoscale complex Eulerian models such as the EMEP Unified model, EMEP4UK (Vieno et al., 2009a), and CMAQ (Byun and Schere, 2006) have much longer run-times per simulated year; for a domain and resolution of similar size and resolution as the one used here. An annual simulation for FRAME can be completed in ~20 min (100 processors cluster), making it possible to conduct multiple simulations in a useful time for policy applications. Furthermore the short run time makes it feasible to apply this type of model to integrated assessment modelling (Oxley et al., 2003), requiring hundreds of simulations and uncertainty studies typically requiring thousands of simulations (Page et al., 2004).

Eulerian models are, as mentioned above, very computationally expensive; however, they can be used to simulate episodes of short term air pollution events (hourly) as well as annual averages. As an example the EMEP4UK has been used to simulate the evolution in August 2003 of high ozone concentrations over the UK on an hourly time scale (Vieno et al., 2009b). However, even with recent advance in computer power, the large set of runs needed (for example for uncertainty analysis) still presents a major challenge to Eulerian models.

2. Description of the FRAME model

The FRAME (Fine Resolution Atmospheric Multi-pollutant Exchange) model (Singles et al., 1998) is an Atmospheric Transport-chemistry

Model (ATM) used to simulate annual mean deposition of reduced nitrogen (NH_x = NH₃ + NH₄⁺), oxidised sulphur (SO_x = SO₂ + SO₄²⁻) and oxidised nitrogen (NO_y = NO + NO₂ + NO₃⁻ + HNO₃ + PAN).

FRAME is a statistical Lagrangian model that uses a domain of the United Kingdom and Eire, and is divided into 172 × 244 grid squares of a 5 × 5 km² resolution (Vieno, 2006). The model advects an air column (2.5 km height and area 5 × 5 km²) using a statistically derived wind-speed and wind-frequency along straight trajectories (Dore et al., 2006a). This approach provides a computational advantage compared with more detailed Eulerian models, such as EMEP4UK, enabling simulation of multiple footprints and analysis of abatement scenarios and the individual source contributions to different ecosystem receptor sites.

The air column of FRAME is divided into 33 layers of thickness, varying from 1 m for the lowest layer up to 100 m for the top layer. There are four main processes simulated within the air column: emission, diffusion, chemical reactions and deposition processes. Emissions are injected into the appropriate vertical layers in the model. Dry deposition for ammonia is parameterised using a model of canopy resistance specific to land cover (Singles et al., 1998); spatially variable pre-calculated land-dependent deposition velocities are used for SO₂ and NO₂ (Smith et al., 2000). The remaining species (aerosol and PAN) have a fixed deposition velocity. Dry deposition removes pollutants at the ground from the surface layer of the model (1–2 m).

Wet deposition is parameterised by a scavenging coefficient for each species to calculate the amount of pollutant removed from the air column by rain and also incorporates a seeder–feeder process (Fowler et al., 1988 and Dore et al., 2006b) of the orographic enhancement of wet deposition (Fournier et al., 2005). 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 FRAME model makes no differentiation between in-cloud and below-cloud process and uses an average value of scavenging coefficient to describe the overall effect. Over high rainfall areas, a proportion of the annual precipitation is designated as ‘orographic precipitation’ (Fournier et al., 2005). It is assumed that this excess orographic rainfall is due to an altitudinal effect, and thus a fraction of pollutants are also removed from hill clouds by the seeder–feeder effect. The scavenging coefficient is calculated by assuming that the orographic rainfall will remove material twice as efficiently as non-orographic precipitation. FRAME incorporates the directional dependence of orographic rainfall by considering two components of rainfall: non-orographic precipitation which has no directional dependence and orographic precipitation which is directionally dependent and stronger for wind directions associated with humid air masses. The directional orographic rainfall model is described in detail in Fournier (2002) and Fournier et al., 2005.

The chemical scheme in FRAME is similar to that used in the EMEP Lagrangian Acid Deposition Model (Barret et al., 1995). Oxidised nitrogen chemistry includes NO₂ dissociation, PAN (peroxyacetyl nitrate) formation, NO by ozone reaction and formation of nitric acid. The equilibrium of nitric acid and ammonia controls the formation of ammonium nitrate; ammonium sulphate is produced by reaction of sulphuric acid and ammonia. The aqueous phase reactions considered in the model include the oxidation of S(IV) by O₃, H₂O₂ and the metal catalysed reaction with O₂. The prognostic chemical variables in the model include gas and aerosol concentrations of: NH₃, NH₄NO₃, (NH₄)₂SO₄, NO, NO₂, NO₃⁻, HNO₃, PAN, H₂SO₄ and SO₂, for each model layer, where NO₃⁻ is coarse-mode particulate nitrate.

The parameterisation used in the FRAME model for the plume rise of elevated sources, is as follows (Hanna et al., 1982):

$$\Delta h = \frac{E}{\bar{u}^a} \quad (1)$$

where Δh is the plume rise, E and a are parameters defined below and \bar{u} is the mean wind speed.

Buoyancy forces generally dominate over inertial forces during plume rise when $\Delta T > 50$ K (Seinfeld and Pandis, 1997). All high stack emissions used in the FRAME model have an exit temperature at least 50 K above the ambient temperature, therefore justifying this parameterisation. The parameters E and a are defined for the neutral and unstable conditions in Eq. (2) and for stable conditions in Eq. (3) (ASME, 1973):

$$E(\text{neutral and unstable}) = 7.4 \cdot \left(\frac{g \cdot d^2 \cdot V_s \cdot (T_s - T_a)}{4 \cdot T_s} \cdot h_s^2 \right)^{1/3}, \quad a = 1 \quad (2)$$

$$E(\text{stable}) = 29 \cdot \left(\frac{d^2 \cdot V_s \cdot (T_s - T_a)}{4 \cdot \frac{T_s}{T_a} \cdot \frac{\partial \theta}{\partial z} \cdot (p/p_0)^{0.29}} \right)^{1/3}, \quad a = \frac{1}{3} \quad (3)$$

where g is the acceleration due to gravity (9.81 ms^{-2}), d [m] the stack diameter, V_s [m s^{-1}] the exit velocity, T_s [K] the exit temperature, T_a [K] the ambient temperature, h_s [m] the stack height, p [hPa] the atmospheric pressure, $p_0 = 1013$ hPa, θ [K] is the potential temperature, and z [m] the vertical coordinate.

3. Model application

Model simulations were conducted for a 2003 base year and a future 2010 emissions scenario based on the Gothenburg protocol. The future year simulation consisted of identical meteorological parameter but a different emissions inventory.

The performance of the FRAME model 2003 simulations was assessed by comparison with annual averaged observations for the year 2003 of: SO_2 concentrations, particulate SO_4^{2-} concentrations and wet deposition of sulphate in the UK. The networks used are: the UK Rural SO_2 Monitoring network, which uses fortnightly filter pack sampling (until 2003, www.airquality.co.uk) and the Nitric Acid Monitoring Network which employed the DELTA system (DENuder for Long Term Atmospheric sampling; Sutton et al., 2001) at 12 sites in the UK to make measurements of HNO_3 , SO_2 , NH_3 and aerosol concentrations (Tang et al., 2007). 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. The UK national monitoring networks for air quality and acid deposition have been summarized by Hayman et al. (2004). The FRAME model calculates annual averaged values, therefore the observations of each network are averaged to yearly values.

The parameterisation of plume rise used in the FRAME model requires stack parameters that are not included into the UK National Atmospheric Emissions Inventory (NAEI), where only source magnitude and location are provided. A detailed inventory of emission estimates for individual point sources, for the year 2003 and a 2010 emissions scenario, was therefore assembled based on data provided by the Environment Agency of England and Wales, the Scottish Environmental Protection Agency and the Environment and Heritage Service of Northern Ireland. The assembled database included all the major high-stack point sources for the years 2003 and 2010, comprising all the stack parameters required by the plume rise module used in the FRAME model. At the time of this study the UK NAEI estimates for the remaining gridded emissions were available for the year 2002, therefore the NAEI 2002 non-point source emissions were corrected to the available total NAEI 2003 emissions estimates. This correction had only a minor effect on the overall footprints for individual point sources.

The emissions for the remaining non-point sources includes all Selected Nomenclature for Air Pollution (SNAP) sectors for NH_3 , NO_x and SO_x . Each of the 11 SNAP sectors (Combustion in Energy Production & Transformation, Combustion in Commercial, Institu-

tional & Residential & Agriculture, Combustion in Industry, Production Processes, Extraction & Distribution of Fossil Fuels, Solvent Use, Road Transport, Other Transport & Mobile Machinery, Waste Treatment & Disposal, Agriculture Forestry & Land Use Change, Nature) are injected at fixed heights representative of each sector.

The FRAME model was used to estimate the sulphur deposition footprint (wet and dry) over the United Kingdom, for the 69 largest SO_x point sources included in the UK emissions inventory. A selection of the results is illustrated in Table 1, which reports the plume characteristics and estimated plume rise for the 4 largest point sources in the UK inventory. In the appendix of this paper Table A1 shows the stack characteristic for all the remaining sources. The four largest sources are Cottam (Nottinghamshire, England), West Burton (Nottinghamshire, England), Longannet (Clackmannanshire, Scotland) and Eggborough (North Yorkshire, England), which together account for ~30% and ~15% of the total 2003 and projected 2010 UK SO_2 point source emissions, respectively.

The method used to calculate the deposition footprint for each source consists of running a model baseline simulation for the year 2003 and 2010 and then a series of simulations, each with a different point source removed. The source deposition 'footprint' was then calculated for each grid square of the model domain from the difference between the baseline simulation and the simulations with individual point source emissions removed. This method ensures that appropriate background chemical concentrations, necessary to drive atmospheric gas to particle conversion rates, are employed for all model simulations.

This work was a part of a larger project (Dore et al., 2007b) in which the UK total deposition was reconstructed by adding up all 69 foot prints included here plus a foot print for all the remaining emissions. We found a difference of ~25–30% of total SO_x deposition between the base run (which included all sources) and the sum of all foot prints. To deal with this issue of non-linearity we normalised each foot print to ensure that the sum of the footprints matched the results of the base case. However, the work presented here was not intended to reconstruct the total deposition for 2003 by source but to focus on deposition from individual point sources. This problem has been addressed in previous works by Alpert et al. 1995, Doraiswamy et al. (2007), and Pun et al. (2002).

Table 1

Physical parameters and emissions magnitude for the four largest SO_2 point sources (according to 2003 emissions) included in this study.

Name x,y OS grid (km)	Stack height (m)	Stack diameter (m)	Exit velocity (m s^{-1})	Exit temperature (K)	Example of injection height due to plume rise for a typical values of atmosphere + stack height	Neutral and unstable/ stable (m)	Injection layer in FRAME
Cottam 481.2,379.5	198	12.2	25.6	408	661/ 547		15th/14th
West Burton 479.1,385.6	183	8.9	27.0	403	541/ 468		14th/14th
Longannet 292.5,688.0	183	7.6	18.5	398	464/ 407		13th/13th
Eggborough 457.8,425.4	198.5	12.2	27.2	403	667/ 551		15th/14th

In the FRAME model, each of the ~500 point source is labelled with a unique index. The sources have been sorted by emissions strength for year 2003. The x and y coordinates are in kilometres on the British national grid projection. The effective emission height 'injection height' on the model is shown both in m above ground and as the model layer (the total number of layers in FRAME is 33).

Table 2
FRAME model UK deposition budget for the year 2003 and 2010.

	NH _x Gg Nyr ⁻¹	NO _y Gg Nyr ⁻¹	SO _x Gg Syr ⁻¹
<i>2003 FRAME (CBED 2001–2003) budget (UK)</i>			
Import	36.4	67.1	56.7
Emissions	250.6	445.5	524.0
Dry deposition	62.9 (65)	39.6 (98)	58.3 (64)
Wet deposition	84.0 (107)	52.3 (95)	108.3 (117)
Export	125.4	408.8	390.9
<i>2010 FRAME budget (UK)</i>			
Import	34.4	52.0	49.9
Emissions	239.3	352.5	368.2
Dry deposition	61.0	31.4	42.2
Wet deposition	78.9	41.4	89.4
Export	119.8	322.3	268.3

Within bracket and in bold the 2001–2003 averaged observations derived estimate from the CEH CBED model.

4. Results

A summary of the FRAME model UK budgets for NH_x, NO_y and SO_x for the years 2003 and 2010 are shown in Table 2. Dry and wet deposition of SO_x for each of the 69 point sources are summarised in Table A2 of the Appendix. As comparison the Centre for Ecology and Hydrology (CEH) CBED (Concentration Based Estimated Deposition, Smith et al., 2000) model predicted NH_x, NO_y and SO_x dry and wet UK deposition for the year 2003 is shown in bold and within brackets in Table 1. CBED is an observation derived model which can be used to verify the UK total deposition budget calculations calculated with FRAME.

The FRAME simulations for 2003 of surface SO₂ concentration, SO₄ concentration and wet deposition of SO₄ versus observations are shown in Fig. 1(a,c,d), where all these model runs were made using the plume rise module and actual stack conditions. The model simulated very well the annual surface concentration of SO₂ (Fig. 1a) with a slope of 1.02 and R² of 0.97, but the model slightly

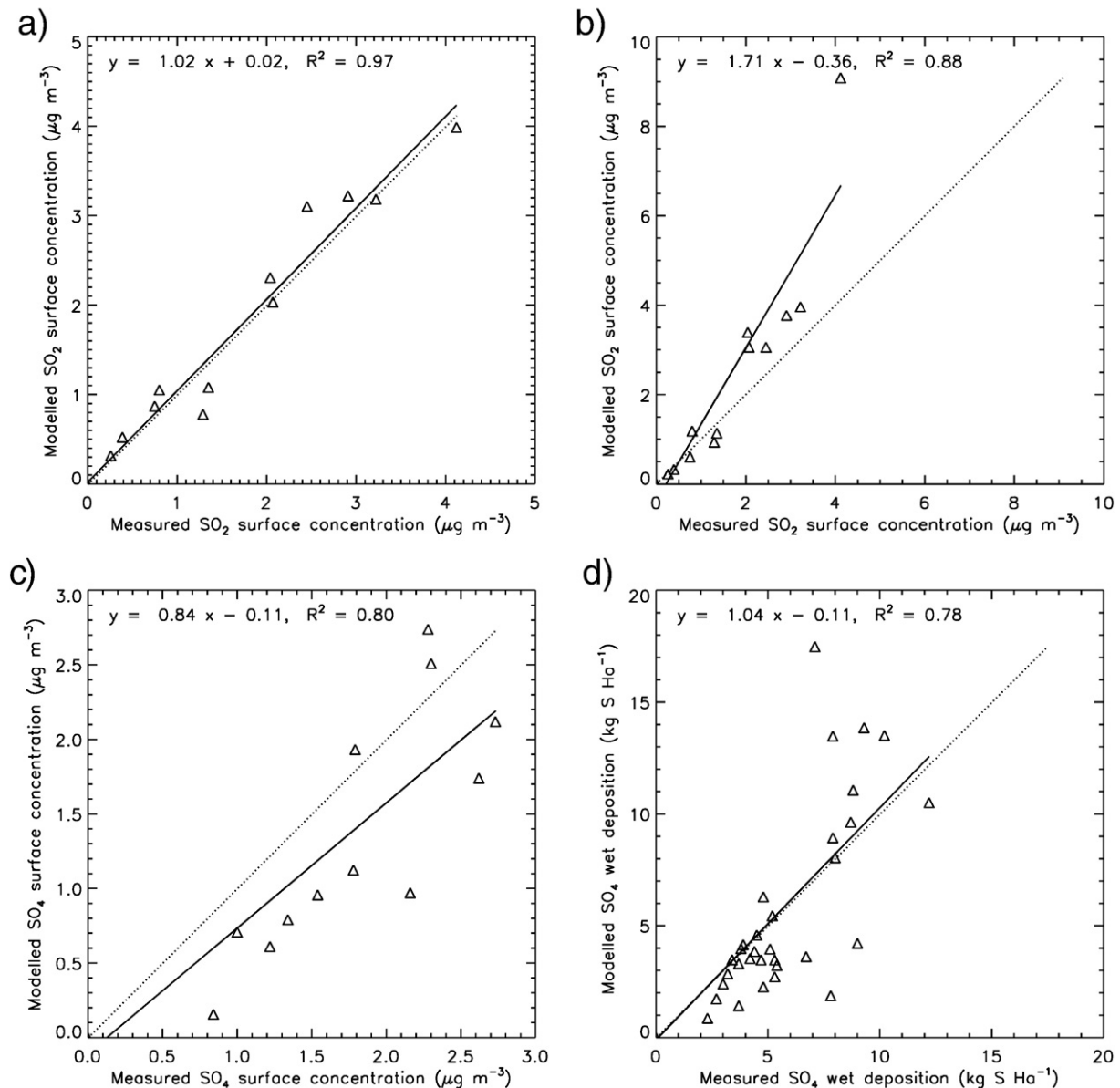


Fig. 1. Comparison of the FRAME model simulations for 2003 with surface air monitoring data: a) SO₂ simulated with the plume rise of elevated sources b) SO₂ simulated without the plume rise of elevated sources. c) SO₄ surface concentration and d) SO₄ wet deposition particulate, both including the effects of plume rise.

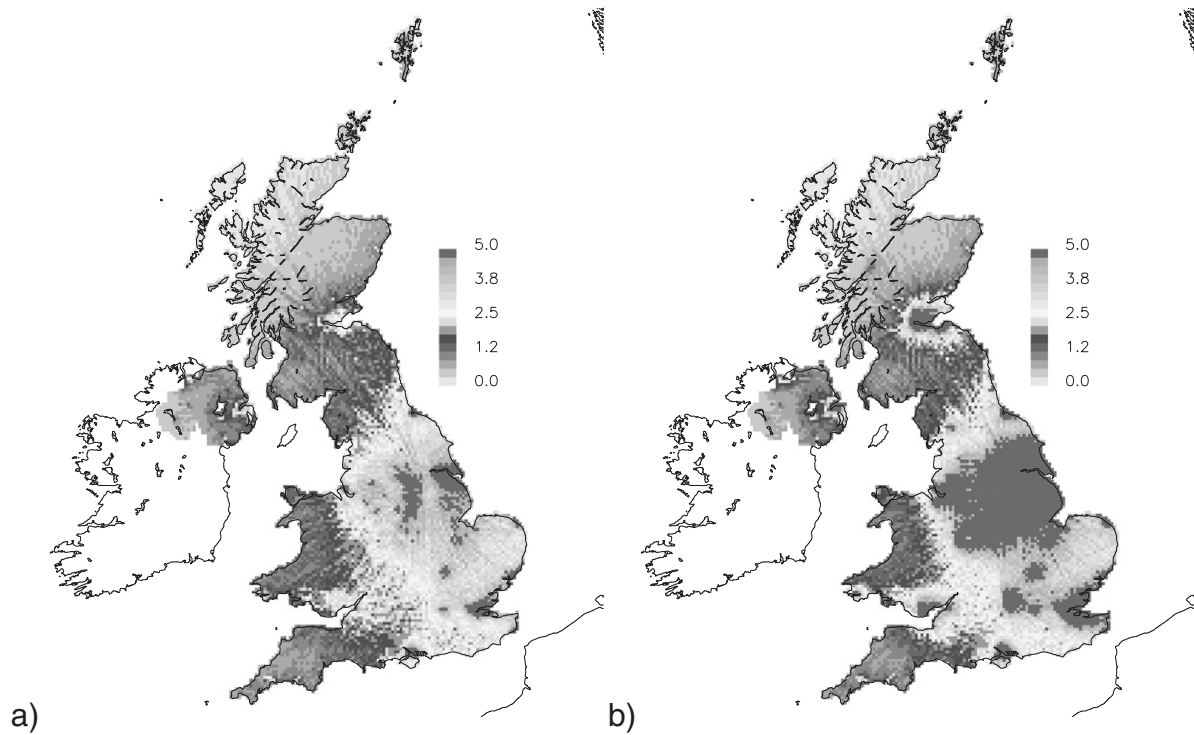


Fig. 2. FRAME model estimated near surface 2003 SO₂ concentrations (layer 1–2 m above ground): a) simulated with plume rise for the major elevated point sources and b) using no plume rise. Units are µg m⁻³. The maps are shown red when concentrations exceeded 5 µg m⁻³. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

underestimated the measured surface concentration of SO₄ (Fig. 1c) with a slope 0.84 and R² 0.80. Annual wet deposition of SO₄ (Fig. 1d) is also well reproduced by the model with a slope of 1.04 and R² of 0.78.

Stack parameters play a key role in accurately representing atmospheric sulphur concentrations. Thus when the model is run without plume rise simulation (i.e., the pollutant is injected at the stack height) for

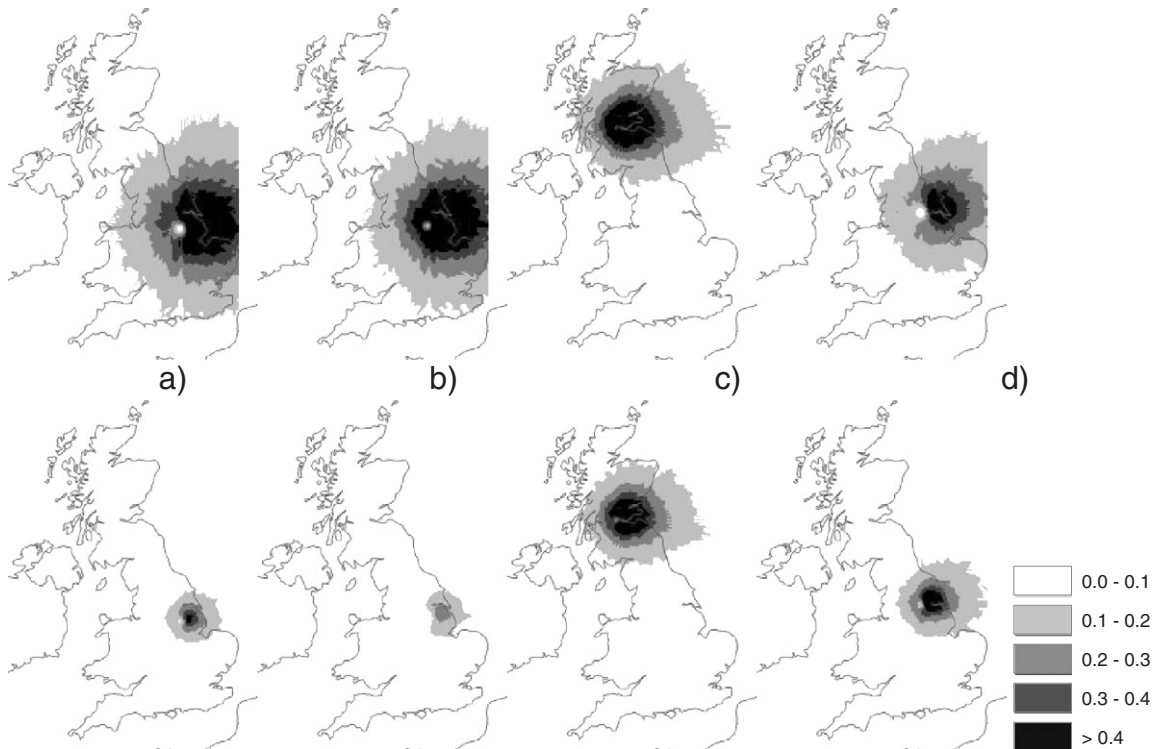


Fig. 3. Footprints of SO_x dry deposition for the four strongest SO_x point sources in the UK as calculated by FRAME. Top: 2003, Bottom 2010: a) Cottam, b) West Burton, c) Longannet and d) Eggborough. Units are kg Sha⁻¹ yr⁻¹.

the elevated point sources, the model over-estimated the measured SO_2 surface concentration by a factor of ~ 1.7 (Fig. 1b). The spatial pattern of SO_2 concentration is also influenced by the plume rise. Fig. 2 shows the FRAME simulated surface concentration of SO_2 : a) with the plume rise and b) without the plume rise. The area of the UK where the concentrations of SO_2 exceed $5.0 \mu\text{g m}^{-3}$ is larger (by a factor ~ 10) when the plume rise of high stack point sources is not used when compared with the model results when the plume rise module is applied. Significant, though smaller, effects of the plume rise module were noted for the simulation of sulphate wet deposition and aerosol concentrations.

Dry deposition of SO_x footprints, for the four largest elevated point sources, are shown in Fig. 3. A recent study by Dore et al. (2006a) quantified the extent to which the annual average wind-rose for the UK has a predominant west-south-westerly wind direction. The effect on the dry deposition footprint is to have an elliptical shape orientated with the predominant wind direction as shown in Fig. 3. At the location of the elevated point source a circle of relatively low dry deposition is visible in all SO_2 footprints. The model simulated diameter of the dry deposition footprint 'eye' is typically 10 ± 5 km, and varies depending on the stack height, stack diameter, temperature and exit velocity of the emitted gases.

In contrast to dry deposition, where the size of the footprint was closely linked with the magnitude of the source, the pattern of the wet deposition is strongly linked to the location of the source, as shown in Fig. 4. Two main factors control this: first, wet deposition is closely linked with rainfall which is higher over upland terrain due to orographic precipitation; secondly, the seeder-feeder mechanism enhances the wet deposition even further (Fowler et al., 1988; Dore et al., 1992, 2006b). As a result wet deposition footprints of point sources located within or near upland terrains may be more intense than stronger point sources located in flatter landscapes. For example, the Longannet point source in central Scotland has a more intense wet deposition footprint with an associated higher amount of sulphur wet

deposited (per unit emission) within the UK than Cottam point source of Nottinghamshire. The location of the 4 sources is shown in Fig. 5c.

The ratio between dry and wet SO_x deposited within the UK from each of the 69 point sources analyzed in this study reflects the location of the source, as particularly affected by the extent of SO_x wet deposition and SO_x exported from the UK, with SO_x dry deposition contributing the largest fraction in the south and east of the UK, but with strong local variation reflecting the emission characteristics of each point source. These differences are shown in Fig. 5a and b for the years 2003 and 2010, respectively.

The differences in stack characteristics, consequent SO_x ratio of dry to wet UK deposition for each stack, and the location in the country, strongly affect the percentage of SO_x emissions that are re-deposited as dry + wet SO_x within the UK from each source, versus the fraction that is exported. This is clearly demonstrated in Fig. 5c and d, which relate the % of emitted SO_x , re-deposited as dry + wet SO_x within the UK to the point source SO_x emission magnitude, for 2003 and 2010. In general, the smallest percentage deposited in the UK (the largest export) is associated with the largest point sources, especially where these occur to the east of the country. However, it is worth noting that the percentage of SO_x estimated to be deposited within the UK generally increases between 2003 and 2010 for the same sites and stack characteristics, highlighting the role of chemical non-linearities related to SO_x emissions reduction.

In both years a cluster of point sources having SO_x dry deposition higher than SO_x wet deposition is present in central England, whereas point sources with higher SO_x wet deposition are generally located closer to hill terrains. It is notable that point sources located in the western part of the UK contribute significantly more to wet deposition than to dry deposition (by factors of 2–4) as air masses passing over these sites during conditions of predominating south-westerly flow are subjected to high rainfall over the hills of Wales, the Pennines in England and the Scottish Highlands. The comparison between the two

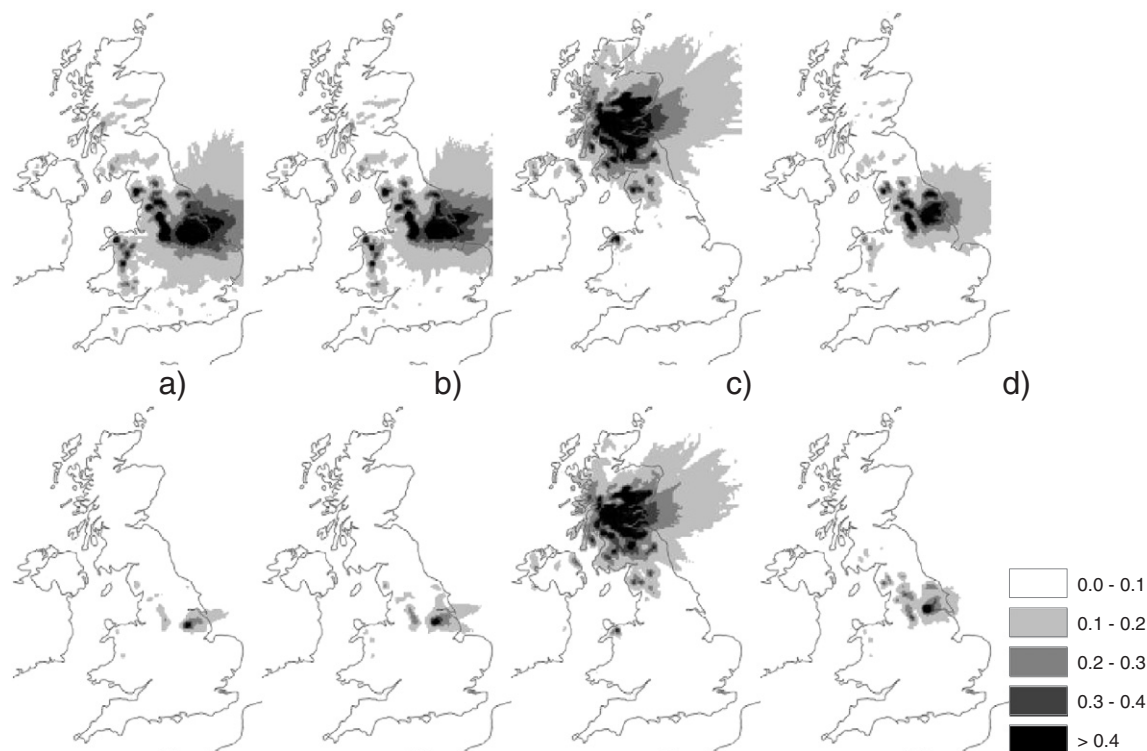


Fig. 4. Footprints of SO_x wet deposition for the four strongest SO_x point sources in the UK as calculated by FRAME. Top: 2003, Bottom 2010: a) Cottam, b) West Burton, c) Longannet and c) Eggborough. Units are $\text{kg S ha}^{-1} \text{yr}^{-1}$.

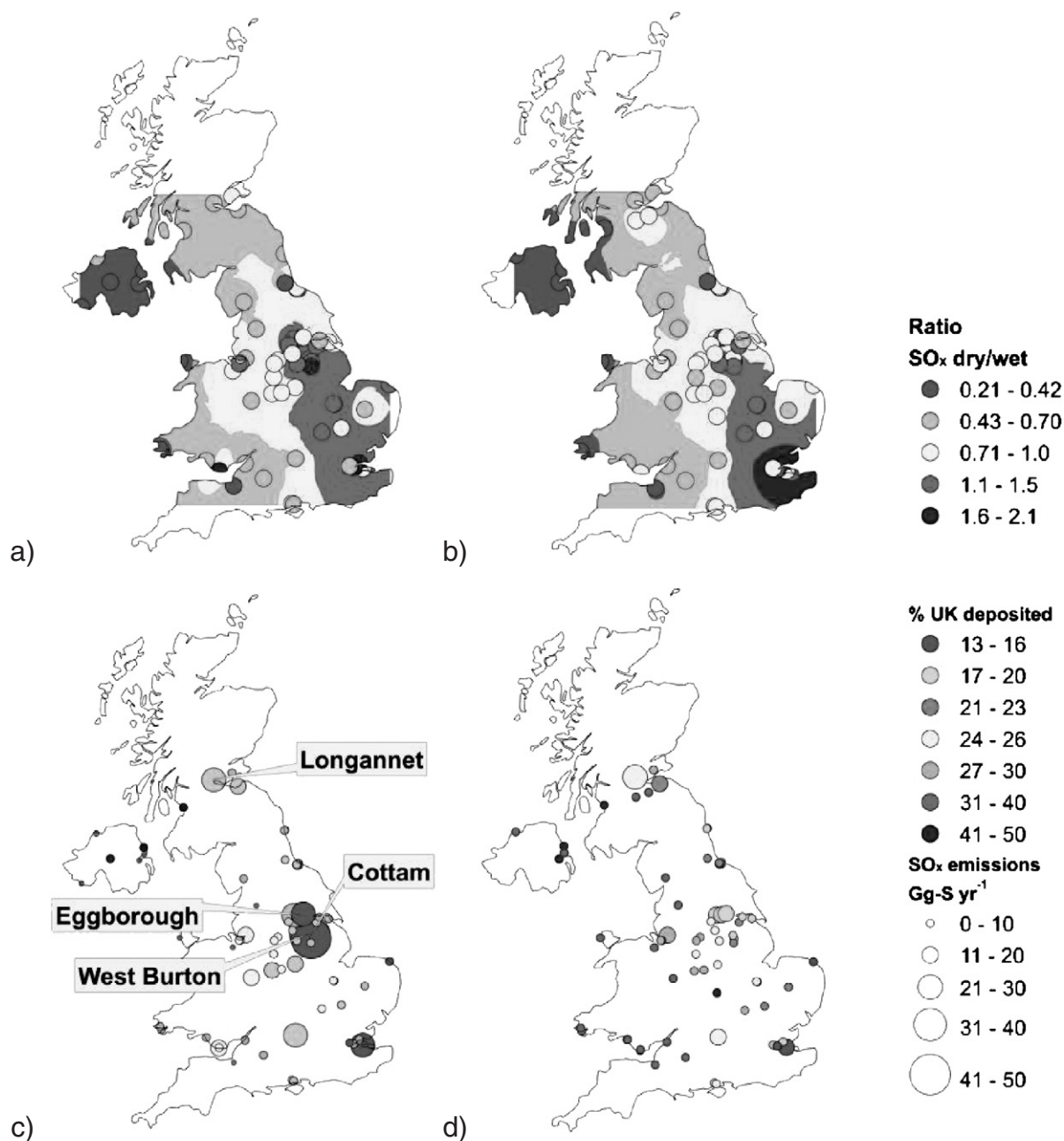


Fig. 5. Deposition characteristics each of the major SO_x point sources in the UK. Top: Ratio plots of SO_x dry/wet deposition for each SO_x point source, for a) 2003 and b) 2010. An interpolated field of the point source dry/wet ratio is also shown to better visualise the ratio spatial pattern. Bottom: Percentage of SO_x emission for each point source deposited as total SO_x (dry + wet) within the UK, for c) 2003 and d) 2010. In the bottom maps, the circle size indicates the magnitude of the SO₂ emission (Gg Syr⁻¹) for each elevated point source. Location of the four 2003 major sources is also shown.

simulated years shows in general a reduction of the ratio of dry to wet deposition (Fig. 5a, b). This occurs because a larger fraction of the emitted SO₂ is converted to sulphate aerosol in 2010 as oxidation proceeds more freely in a generally lower SO₂ climate with oxidant depletion being a less significant factor. This changing in the atmospheric chemistry over the UK, between 2003 and 2010, is illustrated in Fig. 6 which shows the ratio of modelled concentration of SO₂/SO₄ for the two years.

5. Discussion and conclusions

A Lagrangian atmospheric transport model was applied to simulate SO_x emission, dispersion and deposition over the United Kingdom. The

introduction of a dataset including all stack parameters the 69 largest point sources allowed the application of a plume rise module for each point source, leading to a much improved correlation with measurements of ground level SO₂ concentrations.

It can be concluded therefore that accurate representation of high stack point source in an atmospheric transport model is critical in order to obtain good model correlation with measurements of gases that have hot buoyant emissions from elevated stacks. Given the wide range of stack characteristics and their geographical variation, it can be concluded that explicit site-based representation of SO₂ plume rise should be included into all ATMs used for policy applications. Although the plume rise is a well understood atmospheric process, it is not always included in regional ATMs. In addition, in some cases,

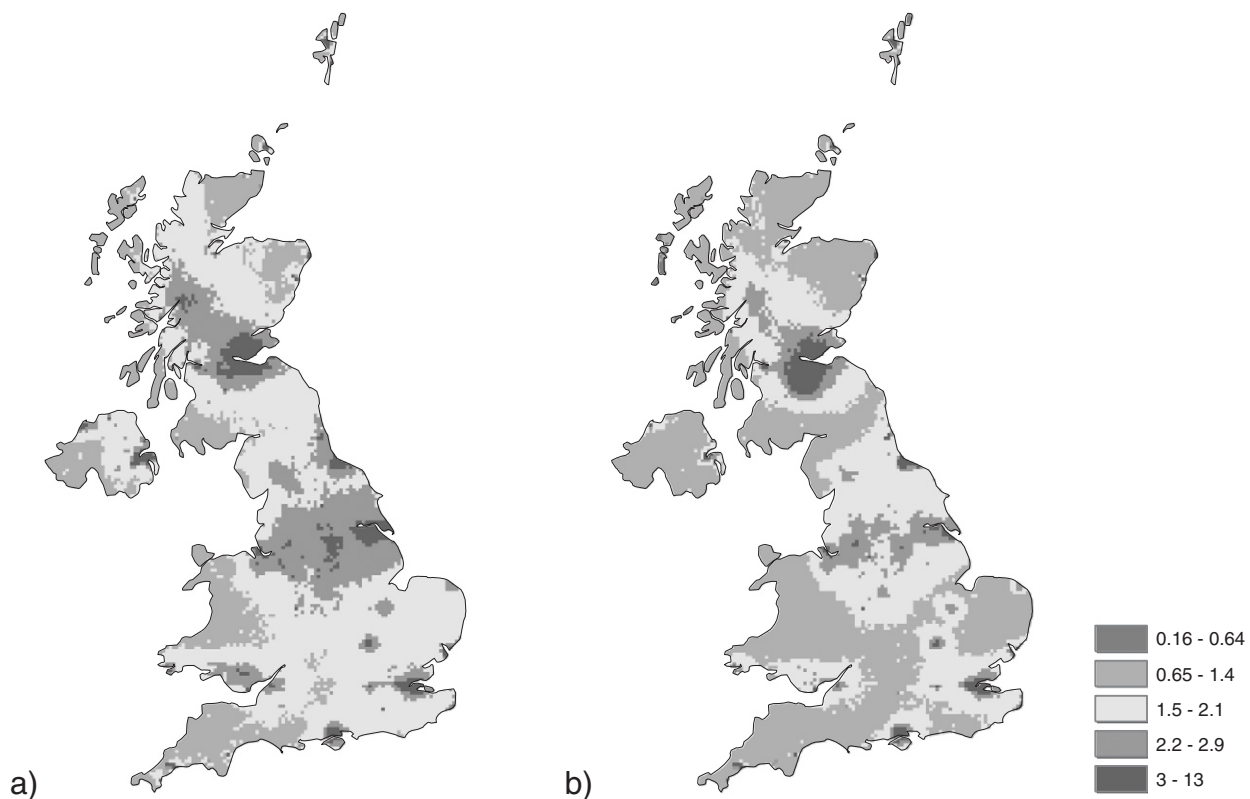


Fig. 6. Ratio plot of the FRAME model calculated concentration of SO_2/SO_4 : a) 2003 and b) 2010.

only default values of stack parameters are used, which may introduce significant errors to effective stack heights. As an example, the EMEP Unified model may be considered, which is used to assess sulphur, nitrogen and ozone deposition over Europe under the UNECE Convention on Long Range Transboundary Air Pollution. This model, does not include plume rise of elevated sources, but instead applies a fixed vertical distribution of the main sources (Simpson et al., 2003). Such an approach represents a practical solution to the difficulty in obtaining stack parameter for European point sources. However, as is clearly illustrated here (e.g. Figs. 1, 2 and 5), simulation of plume rise related to individual stack characteristics has a major influence on the model performance, both in terms of estimated ground level concentrations, and in regard of the fraction exported for each source. For this study in the UK, it has proved possible to obtain the stack characteristics for each of the main point sources, and it should thus be a priority to obtain and apply the equivalent data in models for other regions.

The FRAME model well simulated the UK national deposition budget when compared with the CBED measurement-based estimates. SO_x dry and wet deposition are well reproduced by the model and are within 10% of the CBED estimates. Reduced nitrogen (NH_x) also agrees well with CBED, however a significant discrepancy is present for oxidised nitrogen (NO_y). In CBED the largest part of total NO_y deposition is from HNO_3 . There is some evidence that FRAME underestimate surface concentration of HNO_3 (not shown here) and therefore deposition. However, uncertainty in dry deposition with CBED needs to be taken in account as the HNO_3 national monitoring network in 2003 comprised only 12 sites. This resulted in high uncertainty in the CBED mapped concentrations of HNO_3 across the UK obtained by interpolation.

The FRAME model has also here been applied to illustrate the dry and wet deposition footprints of sulphur associated with the four

largest point sources in the UK (Figs. 3 and 4). The role of source magnitude and stack characteristics are clearly shown in the dry deposition footprints. For example, Table 1 shows that Longannet had a smaller stack diameter, exit velocity and exit temperature than the three other largest sources. This led to a smaller calculated plume rise and less well defined 'eye' immediately adjacent to the source (Fig. 3c). For wet deposition, the proximity of a source to high precipitation upland areas played a large role in determining the extent of the areas with high deposition. Additionally, the magnitude of the emissions source was important, and again this is particularly clearly indicated by the footprint for Longannet (Fig. 4c). Point sources which have weaker emissions, but are located near mountains may deposit the same or similar, amount of sulphur to the UK as compared to stronger sources which are located in the lowlands. With possible climate change the UK rainfall may increase (IPCC, 2007), making wet deposition even more important.

Location of the point sources also influences the fraction of pollutant exported outside of the UK. In Fig. 5c and d for 2003 and 2010, respectively, a clear gradient west to east is visible for the % of emitted SO_x re-deposited within the UK as SO_x . The predominant SW wind direction directly advects pollutants outside the UK more efficiently if the point source is located in the east side of the UK. Another cause of the west–east gradient is the location of hilly terrain mainly located on the west side of the UK. The policy implication of these results is the possibility of ranking the most efficient way to reduce UK SO_2 emissions and sulphur deposition to meet environmental targets.

The use of these results in an Integrated Assessment (Oxley et al., 2003) will have implications for specifying and assessing policy scenarios in relation to the spatial location of power generation (and stack heights of individual power stations) depending upon whether the policy is targeted at protecting

national ecosystems, air quality or reducing transboundary export. A discussion of emission reduction based on an optimisation routine that iteratively combines deposition foot-prints and critical loads maps to minimise critical loads exceedance can be found in Whyatt and Metcalfe (2004).

A key message is that as the emissions are reduced the percent that is exported is also reduced (Fig. 5d). The consequence is that with lower emissions in 2010 compared with 2003, national deposition from UK SO_x emissions does not decrease in proportion to the emission reductions. Thus SO_x emissions plus SO_x import are estimated to decrease by 28% between 2003 and 2010, while modelled total deposition decreased by 21% (Table 2). Conversely, exports of UK SO₂ emissions reduce more than proportionally to the emission reductions (31%). Overall, the % of emissions re-deposited within the UK for 2003 and 2010 is 32% and 36%, respectively. This non-linearity may be explained by a higher rate of SO₂ oxidation in a chemical climate with lower SO₂ concentrations. (Table 2). This is illustrated by the changing ratio of the concentration of SO₂/SO₄ (Fig. 6) between 2003 and 2010. Under conditions of large emissions from a point source oxidant depletion will constrain the SO₂ to remain in the gas phase longer. With reduced SO₂ emissions, a larger fraction of the SO₂ is oxidized to SO₄ in the model simulation (Fig. 6), allowing a larger fraction to be wet deposited over the UK. In parallel, as most emissions of SO₂ originate from high stacks, the SO₂ surface concentrations are kept relatively low, so that dry deposition provides a much less significant pathway for removal than wet deposition (Table 2). Although SO₂ is soluble, the aerosols are washed out more quickly because they act as cloud condensation nuclei and are incorporated into solution at the point of cloud formation. This is not explicitly modelled in FRAME, but is the reason for higher washout coefficients for aerosol used in the model (Fournier, 2002). The effect results in less-than-proportional UK benefits of reducing in SO₂ emissions, together with greater-than-proportional benefits in reducing export of UK SO₂ emissions. This effect is consistent with measured trends in SO₂ concentrations and SO₄ deposition (Fowler et al., 2005) and should be taken in to consideration when emissions reduction policies are enforced.

The non-linearity of the concentration ratio of SO₂/SO₄ between the two years included here, is particularly important as ammonia emissions are hard to control. Ammonia emissions from the UK have shown only small decreases in recent years, 350 Gg yr⁻¹ for 1990 with an emissions forecast of ~300 Gg yr⁻¹ for the year 2010. This is ~14% reduction in 20 years. Therefore to control the S deposited to the UK ammonia needs to be taken into account. A multi-pollutant approach needs to be used when emissions reduction policies are created.

In this study, all 69 major SO₂ point sources were investigated using the FRAME model. A simple model such as FRAME, does not have the ability to simulate short-term pollution events, as it uses statistically derived annual meteorological parameters (Dore et al., 2006b). However, the comparison with observation illustrated here proves the model to be a robust tool for estimation of annual average sulphur deposition, which can be used for policy maker support, and applied to uncertainty analysis.

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Appendix A

Table A1
Point sources stack parameters for the 69 point sources included in this study.

Index	Height (m)	Velocity (ms ⁻¹)	Temp (K)	Diameter (m)
1	198.0	25.60	408.00	12.20
2	183.0	27.00	403.00	8.90
3	183.0	18.50	398.00	7.60
4	198.5	27.20	403.00	12.20
5	259.0	21.60	363.00	15.80
6	199.0	29.40	363.00	12.00
7	198.0	26.00	403.00	12.20
8	198.0	24.60	408.00	9.10
9	183.0	22.00	403.00	8.40
10	152.0	25.10	413.00	11.90
11	204.0	27.60	403.00	8.50
12	198.0	17.20	403.00	12.20
13	198.0	17.40	353.00	14.20
14	149.0	22.00	401.00	7.40
15	171.0	19.10	413.00	8.40
16	160	16	423	5
17	137.2	15.70	473.00	5.15
18	200.0	16	393.00	5.57
19	160	16	423	5
20	137.0	16.79	518.00	4.70
21	114.3	19.76	413.00	6.07
22	70.0	9.10	381.00	2.35
23	160	16	423	5
24	96.0	13.29	473.00	3.80
25	160	16	423	5
26	117.0	12.85	446.00	2.60
27	53.0	10.74	423	0.98
28	160	16	423	5
29	71.0	10.00	403.00	2.60
30	120.0	10.10	566.00	1.98
31	79.0	10.00	403.00	2.60
32	160	16	423	5
33	110.0	12.50	408.00	3.35
34	160	16	423	5
35	109.0	16.70	463.00	3.60
36	160	16	423	5
37	70.0	5.00	498.00	6.48
38	73.0	16	423.00	5.38
39	76.5	15.40	309.70	6.70
40	113.8	43.40	589.00	4.57
41	160	16	423	5
42	96.0	17.20	524.00	2.90
43	160	16	423	5
44	160	16	423	5
45	137.0	11.00	403.00	4.88
46	160	16	423	5
47	49.0	5.80	433.00	2.50
48	81.0	16	423.00	5.82
49	160	16	423	5
50	100.6	12.43	389.25	2.34
51	124.0	16	408.00	10.20
52	160	16	423	5
53	122.0	7.50	448.00	3.66
54	160	16	423	5
55	88.4	4.00	523.00	4.27
56	160	16	423	5
57	160	16	423	5
58	73.5	16	418.00	1.84
59	210.0	32.80	428.00	6.50
60	198.0	31.40	427.85	6.10
61	160	16	423	5
62	63.0	13.50	536.00	3.05
63	160	16	423	5
64	160	16	423	5
65	76.2	18.00	493.00	2.30
66	160	16	423	5
67	110.0	15.00	453.00	3.17
68	56.0	22.90	413.00	1.20
69	160	16	423	5

If the stack data were not available a default (*italic*) was used.

Table A2

Deposition within the UK of each of the 69 point sources included in this study for the year 2003 and a 2010 emissions scenario.

Index	Easting (m)	Northing (m)	2003						2010						% Emi reduction
			E (Ggyr ⁻¹)	SO _x DD (Ggyr ⁻¹)	SO _x WD (Ggyr ⁻¹)	DD+WD (Ggyr ⁻¹)	Emi UKdep %	Dry/ wet	E (Ggyr ⁻¹)	SO _x DD (Ggyr ⁻¹)	SO _x WD (Ggyr ⁻¹)	DD+WD (Ggyr ⁻¹)	Emi UKdep %	Dry/ wet	
1	481,200	379,500	43.95	3.63	3.29	6.92	15.75	1.10	7.69	0.77	0.69	1.46	18.99	1.12	83
2	479,100	385,650	40.75	3.72	3.04	6.76	16.59	1.22	7.66	0.66	0.76	1.42	18.54	0.87	81
3	292,536	688,083	26.78	2.71	3.85	6.56	24.49	0.70	21.00	2.08	3.24	5.32	25.33	0.64	22
4	457,830	425,400	25.99	2.10	1.99	4.09	15.74	1.06	12.61	1.27	1.03	2.30	18.24	1.23	51
5	466,100	427,200	22.11	1.63	1.83	3.46	15.65	0.89	15.17	1.08	1.48	2.56	16.88	0.73	31
6	451,000	191,300	21.00	1.82	2.75	4.57	21.76	0.66	17.50	1.61	2.54	4.15	23.71	0.63	17
7	581,159	172,043	20.93	1.10	1.41	2.51	11.99	0.78	17.50	1.01	1.23	2.24	12.80	0.82	16
8	447,600	424,600	20.50	1.91	1.82	3.73	18.19	1.05	17.50	1.56	1.83	3.39	19.37	0.85	15
9	405,600	317,900	19.57	2.15	2.53	4.68	23.91	0.85	8.75	1.18	1.35	2.53	28.91	0.87	55
10	302,400	166,300	18.59	1.93	2.99	4.92	26.47	0.65	5.75	0.63	1.06	1.69	29.39	0.59	69
11	365,350	303,695	18.10	2.03	2.88	4.91	27.13	0.70	8.75	1.01	1.73	2.74	31.31	0.58	52
12	354,400	386,200	12.95	1.27	2.21	3.48	26.87	0.57	17.50	1.89	2.90	4.79	27.37	0.65	-35
13	450,170	330,163	12.32	1.19	1.49	2.68	21.76	0.80	7.90	0.72	1.12	1.84	23.29	0.64	36
14	339,500	675,500	11.45	0.90	1.65	2.55	22.27	0.55	10.71	0.82	1.63	2.45	22.88	0.50	6
15	566,100	175,700	7.95	0.62	0.55	1.17	14.72	1.13	9.19	0.71	0.68	1.39	15.13	1.04	-16
16	350,600	495,880	7.50	0.89	1.51	2.40	32.00	0.59	7.50	0.87	1.58	2.45	32.67	0.55	0
17	444,215	103,925	7.11	0.82	0.90	1.72	24.18	0.91	7.11	0.81	0.95	1.76	24.75	0.85	0
18	157,492	543,108	6.90	0.54	1.93	2.47	35.80	0.28	1.85	0.11	0.64	0.75	40.54	0.17	73
19	343,800	375,800	5.45	0.77	0.82	1.59	29.17	0.94	5.45	0.76	0.86	1.62	29.72	0.88	0
20	515,360	417,770	5.27	0.42	0.54	0.96	18.22	0.78	5.28	0.41	0.58	0.99	18.75	0.71	0
21	430,500	590,400	4.67	0.29	0.53	0.82	17.55	0.55	4.67	0.28	0.54	0.82	17.56	0.52	0
22	501,726	242,642	4.28	0.69	0.54	1.23	28.72	1.28	4.28	0.68	0.59	1.27	29.67	1.15	0
23	191,000	202,300	3.50	0.31	0.84	1.15	32.90	0.37	3.50	0.30	0.86	1.16	33.14	0.35	0
24	491,460	410,290	3.28	0.40	0.30	0.70	21.37	1.33	3.28	0.39	0.33	0.72	21.95	1.18	0
25	480,540	371,040	3.03	0.37	0.22	0.59	19.47	1.68	3.03	0.35	0.29	0.64	21.12	1.21	0
26	574,250	182,480	2.90	0.36	0.17	0.53	18.27	2.12	2.90	0.36	0.17	0.53	18.28	2.12	0
27	632,800	335,000	2.72	0.20	0.19	0.39	14.35	1.05	2.72	0.20	0.21	0.41	15.07	0.95	0
28	424,000	319,500	2.52	0.33	0.35	0.68	26.98	0.94	2.52	0.32	0.41	0.73	28.97	0.78	0
29	525,520	297,155	2.07	0.32	0.21	0.53	25.58	1.52	2.08	0.32	0.23	0.55	26.44	1.39	0
30	455,980	525,860	1.96	0.16	0.20	0.36	18.35	0.80	1.96	0.16	0.20	0.36	18.37	0.80	0
31	524,340	297,750	1.92	0.27	0.21	0.48	25.07	1.29	1.91	0.27	0.22	0.49	25.65	1.23	1
32	188,600	208,700	1.90	0.17	0.45	0.62	32.62	0.38	1.90	0.16	0.47	0.63	33.16	0.34	0
33	329,183	362,205	1.74	0.28	0.36	0.64	36.79	0.78	1.73	0.27	0.39	0.66	38.15	0.69	1
34	277,600	186,600	1.67	0.19	0.40	0.59	35.24	0.48	1.67	0.18	0.43	0.61	36.53	0.42	0
35	375,053	443,621	1.48	0.20	0.32	0.52	35.16	0.63	1.48	0.19	0.34	0.53	35.81	0.56	0
36	416,800	382,300	1.45	0.18	0.21	0.39	26.88	0.86	1.45	0.18	0.23	0.41	28.28	0.78	0
37	456,620	525,740	1.43	0.12	0.14	0.26	18.24	0.86	1.43	0.12	0.14	0.26	18.18	0.86	0
38	147,879	531,767	1.28	0.14	0.33	0.47	36.74	0.42	1.28	0.12	0.41	0.53	41.41	0.29	0
39	429,300	589,800	1.23	0.09	0.14	0.23	18.65	0.64	1.27	0.09	0.15	0.24	18.90	0.60	-3
40	452,400	523,400	1.20	0.09	0.13	0.22	18.33	0.69	1.20	0.09	0.13	0.22	18.33	0.69	0
41	353,440	181,630	1.16	0.13	0.26	0.39	33.57	0.50	1.16	0.13	0.28	0.41	35.34	0.46	0
42	453,540	375,160	1.14	0.16	0.11	0.27	23.75	1.45	1.14	0.16	0.13	0.29	25.44	1.23	0
43	437,600	412,200	0.96	0.12	0.11	0.23	24.05	1.09	0.96	0.11	0.12	0.23	23.96	0.92	0
44	330,980	138,260	0.90	0.07	0.25	0.32	35.58	0.28	0.90	0.07	0.26	0.33	36.67	0.27	0
45	456,800	522,000	0.84	0.07	0.08	0.15	17.76	0.88	0.84	0.07	0.08	0.15	17.86	0.88	0
46	226,500	381,000	0.75	0.05	0.24	0.29	38.51	0.21	0.75	0.05	0.24	0.29	38.67	0.21	0
47	452,850	523,075	0.70	0.09	0.07	0.16	22.82	1.29	0.70	0.09	0.07	0.16	22.86	1.29	0
48	649,89	584,844	0.70	0.06	0.19	0.25	35.71	0.32	0.30	0.03	0.08	0.11	36.67	0.38	57
49	341,500	375,900	0.69	0.10	0.09	0.19	27.64	1.11	0.69	0.10	0.10	0.20	28.99	1.00	0
50	303,076	167,447	0.67	0.11	0.07	0.18	26.78	1.57	0.67	0.10	0.10	0.20	29.85	1.00	0
51	156,969	556,542	0.65	0.05	0.21	0.26	40.00	0.24	0.25	0.02	0.09	0.11	44.00	0.22	62
52	561,350	175,270	0.64	0.06	0.05	0.11	17.30	1.20	0.64	0.06	0.05	0.11	17.19	1.20	0
53	388,900	152,700	0.63	0.07	0.14	0.21	33.19	0.50	0.63	0.07	0.15	0.22	34.92	0.47	0
54	497,370	421,040	0.52	0.04	0.06	0.10	19.23	0.67	0.52	0.04	0.06	0.10	19.23	0.67	0
55	453,900	521,550	0.45	0.04	0.04	0.08	17.66	1.00	0.45	0.04	0.05	0.09	20.00	0.80	0
56	563,600	174,400	0.35	0.04	0.02	0.06	17.14	2.00	0.35	0.04	0.01	0.05	14.29	4.00	0
57	278,150	186,920	0.35	0.04	0.07	0.11	31.61	0.57	0.35	0.04	0.08	0.12	34.29	0.50	0
58	65,022	583,475	0.33	0.04	0.08	0.12	36.36	0.50	7.28	0.08	0.11	0.19	63.33	0.73	9
59	555,800	176,600	0.33	0.02	0.03	0.05	15.17	0.67	0.63	0.06	0.06	0.12	19.05	1.00	-91
60	447,400	102,200	0.25	0.02	0.04	0.06	24.10	0.50	0.30	0.03	0.04	0.07	23.33	0.75	-20
61	409,620	375,550	0.25	0.03	0.04	0.07	28.40	0.75	0.25	0.03	0.04	0.07	28.00	0.75	0
62	327,700	701,800	0.22	0.03	0.04	0.07	32.56	0.75	0.69	0.08	0.12	0.20	28.99	0.67	-214
63	408,500	349,600	0.20	0.03	0.03	0.06	29.48	1.00	0.20	0.02	0.03	0.05	25.00	0.67	0
64	430,800	532,800	0.19	0.01	0.03	0.04	20.87	0.33	0.19	0.01	0.03	0.04	21.05	0.33	0
65	453,064	523,780	0.18	0.02	0.02	0.04	21.74	1.00	0.18	0.02	0.02	0.04	22.22	1.00	0
66	445,000	395,000	0.17	0.02	0.02	0.04	24.04	1.00	0.17	0.02	0.02	0.04	23.53	1.00	0
67	585,128	286,927	0.15	0.01	0.02	0.03	19.79	0.50	0.15	0.01	0.02	0.03	20.00	0.50	0
68	233,700	634,900	0.10	0.02	0.03	0.05	49.02	0.67	0.29	0.03	0.10	0.13	44.83	0.30	-190
69	539,690	250,510	0.09	0.01	0.01	0.02	22.86	1.00	0.09	0.01	0.01	0.02	22.22	1.00	0

Emissions (E) and deposition (DD dry deposition and WD wet deposition) are shown in Ggyr⁻¹, followed by the % emission deposited within the UK, the overall ratio of dry/wet deposition for the fraction deposited within the UK, and the last column shows the % of emission reduction between 2003 and 2010.

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