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Sulphate trends in Europe: are we able to model the recent observed decrease?

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ABSTRACT

Abundance of sulphate in Europe has decreased substantially during the last two decades. In this paper, we investigate these recent trends in sulphate concentrations by applying the OsloCTM2 model using three different sets of SO_2 emission inventories. We perform time slice model simulations with emissions for the years 1985, 1995 and 2000 and compare our results with observations to investigate if there is consistency between measured and modelled sulphate trends. Overall the model reproduces the levels of sulphur and the decreasing sulphate trends reasonably well, although some discrepancies exist. The model shows a strong reduction in the surface concentration of sulphate similar to the observations, although a slightly smaller decrease. Continental and Eastern Europe experience the largest decrease in sulphate from 1985 to 2000; observations give 65 and 63% decrease, respectively, while modelled decreases are from 42 to 58% depending on the inventory. We have also studied to what extent our model results are sensitive and robust. Based on our model simulations we find that the EMEP emissions of the three sets of emission inventories are best to reproduce the trends in sulphate observations.

1. Introduction

In the 1970s scientists discovered that air pollution was transboundary, i.e. that gases emitted in one country could be transported long distances and deposited in other countries (Grennfelt and Hov, 2005). This knowledge initiated an international collaboration aiming at reducing the emissions of environmental harmful gases, such as sulphur dioxide (SO_2), oxides of nitrogen (NO_x), ammonia (NH_3) and volatile organic compounds (VOCs). A series of international conventions and agreements were negotiated (for instance Convention on LRTAP in 1979, US Canada Memorandum of understanding in 1980). As a result, emissions (and concentrations) of these gases have been significantly reduced in Europe during the last two decades (Fricke and Beilke, 1992; Grennfelt and Hov, 2005).

Hence sulphur is one area where political agreements and international conventions have proved successful. Between 1980

and 2000 the land based emissions of sulphur dioxide in Europe decreased by nearly 70% (Lövblad et al., 2004). Sulphur dioxide emission reductions were largest in Europe in the 1990s. The trend has levelled out, and for some countries increased in this century. The total European emissions were in 2004 for the first time lower than the 2010 ceilings set by the 1999 Multi-effect UN Protocol (Gothenburg Protocol). This does not mean that all the countries which have signed the Protocol has yet reached their targets, and further sulphur emissions are expected by 2010. Projected emissions modelled by The International Institute for Applied Systems Analysis (IIASA) (Amann et al., 2005) shows a continued SO_2 decrease towards 2020 for the EU-25 countries. The ships emissions are however projected to increase in this period.

Sulphur reductions are mostly due to abatement technologies (e.g. Flue Gas Desulphurization processes, FGD), switching of fuel (from coal to gas) and economic recession (in Eastern Europe). Previously the concern about anthropogenic emissions of sulphur was mostly linked to the acid rain problem: the focus is now on climate effects due to sulphate aerosols (Lelieveld et al., 2002). Sulphate is a result of oxidation of SO_2, both in the gas phase (by OH) and in the aqueous phase (by O_3, H_2O_2,

HO$_2$NO$_2$ and metals). Lifetime is of the order of 1–2.5 d for SO$_2$ and 4–6 d for sulphate (Koch et al., 1999; Chin et al., 2000a; Rasch et al., 2000; Rotstayn and Lohmann, 2002; Iversen and Seland, 2002; Berglen et al., 2004). The influence of sulphur is therefore basically regional.

In this paper, we will use our global OsloCTM2 model with sulphur cycle included (Berglen et al., 2004) to study the recent decrease in sulphur emissions in Europe and its effect on sulphate concentrations. The model will use three sets of emission inventories (EMEP, GEIA/EDGAR/AEROCOM (hereafter called GEA) and Smith et al. (2004) representing the years 1985, 1995 and 2000. EMEP emission inventories are elaborated for Europe only, while GEA and Smith et al. are global inventories. The model results will be compared with observations for the same years for Europe. Through these comparisons we will be in a better position to understand sulphate trends in Europe. For example, Mylona (1996) have estimated historical trends in emissions, but we will focus on more recent trends in this study.

Among the issues we want to address is whether we are able to model the recent decrease of sulphate in Europe and validate the emission inventories. Past studies (e.g. Boucher and Pham, 2002) have investigated historical sulphate trends, but we want to focus more specifically on trends in Europe.

2. Approach

2.1. Model description

In this study we use the tropospheric version of the OsloCTM2 model with sulphur chemistry coupled interactively to a detailed 'ozone' chemistry scheme (Berglen et al., 2004). The model is run in T42 horizontal resolution (2.8° × 2.8°) with 40 vertical layers in σ-hybrid coordinates extending up to 43 km. Advection is solved using the second-order moment (Prather, 1986). Eddy diffusion coefficients from Holtslag et al. (1990) are used for boundary layer mixing. The method by Rodhe and Isaksen (1980) is used for dry deposition, wet deposition in convective and large scale clouds are treated separately (Berglen et al., 2004). The QSSA solver (Hesstvedt et al., 1978) is used in the chemistry scheme comprising 51 components in the tropospheric O$_3$–NO$_x$–VOC cycle. In addition, five sulphur components (DMS, SO$_2$, sulphate, H$_2$S and MSA) are calculated online with the oxidants (Berglen et al., 2004). Meteorological input data are produced by the IFS model at the ECMWF, giving very detailed and internally consistent weather data (mass fluxes, cloud properties, T, p, humidity, etc.). These input data are updated every 3 hr. Meteorological input data representing year 2000 is used for all model runs, except where otherwise stated. We have chosen to use the same year throughout to exclude changes in composition due to interannual meteorological variations.

2.2. Emission data

The annual mean for the three sets of SO$_2$ emissions for the three selected years 1985, 1995 and 2000 are given in Fig. 1.

The EMEP (Cooperative programme for monitoring and evaluation of the long-range transmission of air pollutants in Europe) emission inventories (Vestreng et al., 2004) estimated anthropogenic emissions for Europe based on numbers officially reported by each country under the Convention on LRTAP and annually reviewed by an expert panel. The 11 categories used are energy combustion, non-industrial combustion, manufacturing industry, production processes, fossil fuel/geothermal energy, solvent use, road transport, other mobile sources, waste treatment, agriculture and other sources. Vertical distribution is the same as used at MSC-W (www.emep.int/emis2004/table_add_rep.html).

The data set we have named GEA consists of three different global inventories: GEIA 1985 (Benkovitz et al., 1996 and references therein), EDGAR 1995 (Olivier and Berdowski, 2001) and AEROCOM 2000 (Dentener et al., 2006) are all global inventories widely used by the model community. They are assembled by various groups using the best estimates available at the time of selection. These groups have used slightly different approach; GEIA 1985 uses data from EMEP and CORINAIR for Europe, EDGAR 1995 use energy statistics, and AEROCOM 2000 use data from IIASA/RAINS to quantify anthropogenic emissions for Europe. Nevertheless, we think it is appropriate to test these inventories concerning trends since these inventories are the most applied inventories in atmospheric modelling.

Smith et al. (2004) have constructed global seasonal emission inventories for 1850–2000. Emissions are given for nine categories: coal combustion, oil combustion, natural gas, metal smelting, other industrial processes, biomass combustion, land-use, other, ocean bunker fuels, i.e. ships. Emissions are estimated over and under 100 m. Emissions in this inventory are distributed on a global grid based on regional values. For example, while emissions in Western Europe as a whole change over time, the distribution of emissions within Europe does not change. This reflects the intended use of this long-term inventory for global modelling studies.

Emissions from ships are included in the Smith et al. (2004) inventory, for the EMEP and GEA runs we have scaled the Endresen et al. (2003) AMVER inventory for 2000 backwards assuming an annual increase of 1.6%, i.e. that 1985 emissions represent 78.8% and 1995 emissions 92.4% of the emissions for year 2000.

When we study the trends in anthropogenic emissions we must however have in mind that there are also natural emissions of sulphur, such as oceanic emissions of DMS. These emissions are calculated using ocean concentrations from Kettle et al. (1999) and Kettle and Andreae (2000) together with parametrization from Nightingale et al. (2000). H$_2$S, volcanic SO$_2$ and biomass burning of SO$_2$ are all taken from Spiro et al. (1992). All these
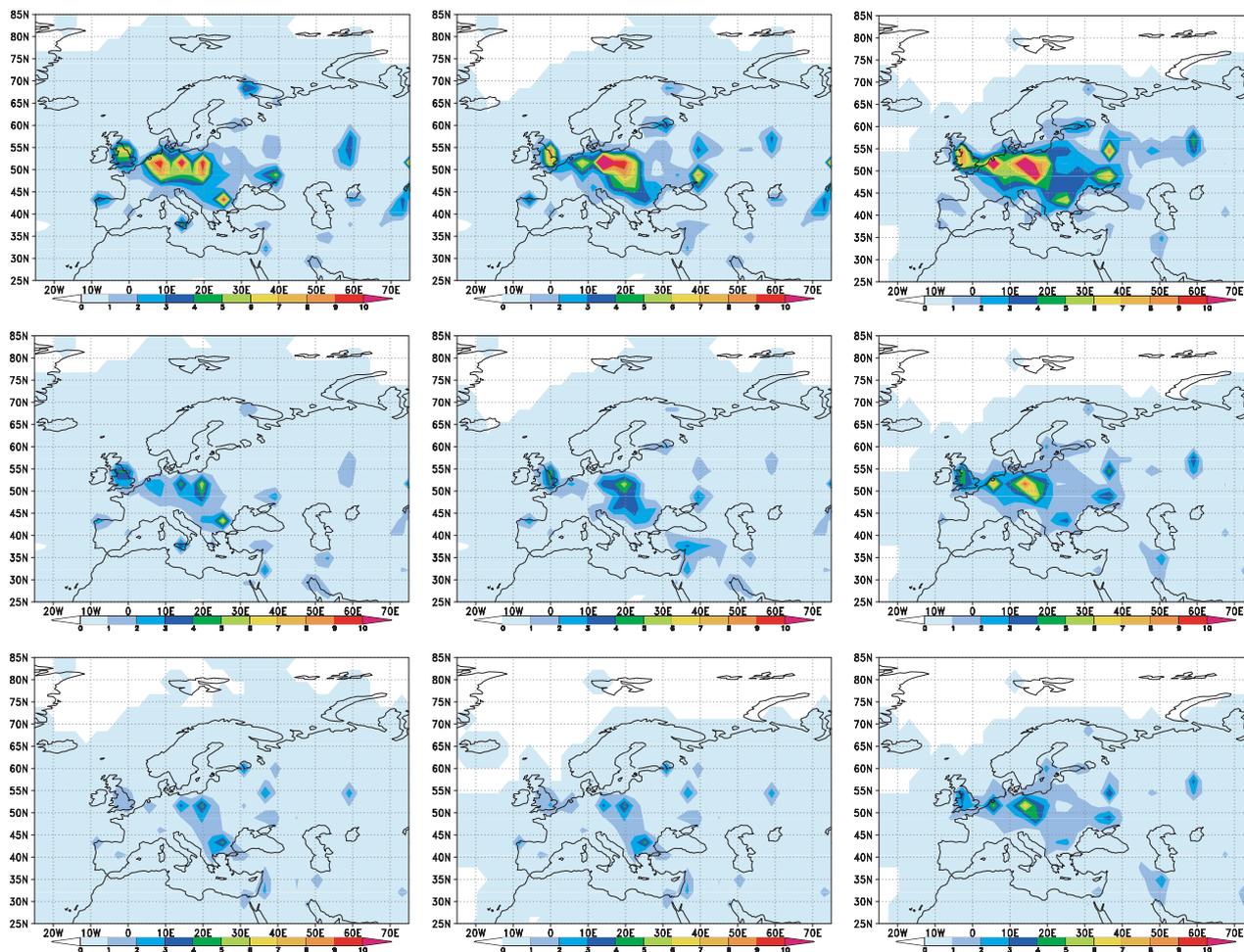
Fig. 1. SO$_2$ emissions, annual mean, 1985 (upper row), 1995 (middle), and 2000 (lower row) using EMEP (left-hand column),
GEIA/EDGAR/AEROCOM (middle) and Smith et al. (right-hand column) emission inventories. Unit: 10^{12} molec m^{-2}s^{-1}.

emissions will provide background concentrations of natural sulphur that does not change over time. In Europe the anthropogenic emissions are much larger than the natural.

2.3. Experimental setup and model runs

Nine model runs were conducted, i.e. three different sets of emission inventories for three different years. We first used $1\frac{1}{2}$ yr of spin-up in T21 (5.6° × 5.6°, 19 layers) with the emission inventory chosen, then 6 months of spin-up in T42 (July–December) and finally 1 yr of model run. Given that tropospheric lifetime of sulphur is on the order of days, this will be more than sufficient spin-up. Also for the oxidants 2 yr of spin-up is considered sufficient for tropospheric purposes (Berglen et al., 2004). An overview of all the different model runs performed is given in Table 1.

We had to make some modifications concerning emissions; EMEP provide only European emissions and other emissions were used elsewhere. However, the impact from intercontinental transport is small compared to the impact from local emissions so the error is assumed to be small. Emissions of oxidants precursors (NO$_x$, CO and hydrocarbons) are adjusted according to the year we run (see Table 1). The signal from changes in emissions of oxidants precursors is small compared to the signal from changes in emissions of sulphur, hence the error introduced by the NO$_x$ and CO emissions is assumed to be small.

2.4. Selection of observations for comparison

To validate our model results we will compare with observations from the EMEP network (Hjellbrekke, 2005). This network organizes observations from all over Europe and assures a common quality standard and format of the observation data. More than 175 stations report or have reported data, of which about 80 monitor or have monitored sulphur components.

When we compare our calculated model concentrations of SO$_2$ and sulphate with observations from one specific year (1985/1995/2000) we compare with all stations available. When

Table 1.   Overview of the nine model runs performed in this study with the various emission inventories used

Name of the run	Sulphur emissions, Europe	Sulphur emissions, rest of the world	Emissions of oxidants
Em85	EMEP 1985	GEIA 1985[a]	GEIA 1985[c]
Em95	EMEP 1995	EDGAR 1995[a]	EDGAR 1995
Em00	EMEP 2000	Aerocom 2000[a]	EDGAR 1995[b]
GEA85	GEIA 1985	GEIA 1985	GEIA 1985[c]
GEA95	EDGAR 1995	EDGAR 1995	EDGAR 1995
GEA00	Aerocom 2000	Aerocom 2000	EDGAR 1995[b]
Sm85	Smith et al. for 1985	Smith et al. for 1985	GEIA 1985[c]
Sm95	Smith et al. for 1995	Smith et al. for 1995	EDGAR 1995
Sm00	Smith et al. for 2000	Smith et al. for 2000	EDGAR 1995[b]

[a]EMEP provide emissions for Europe only.
[b]No emissions of oxidants provided for 2000, use EDGAR 1995.
[c]GEIA 1985 provide some oxidants (NO_x), for the rest we adjust EDGAR 1995 backwards using EDGAR-HYDE so that the percentage change from 1985 to 1995 is the same as from 1980 to 1990 found in EDGAR-HYDE.
All runs use meteorological input data representing the year 2000.

Table 2.   Number of stations in the EMEP area reporting observations for SO_2 and sulphate for different years and combination of years

1985	56
1995	79
2000	69
1985 & 1995	32
1985 & 2000	22
1995 & 2000	55
1985 & 1995 & 2000	21

Fig. 2.   Regions used in this study. See Table 3 for colour codes.

we compare trends, i.e. decrease/increase from one year to another, we use only the stations with observations from the 2 yr which the analysis is performed (see Table 2 for the number of stations used for this comparison). In order to make the comparison of trends more representative for the model domain, we have grouped the stations into six different geographical regions (see Table 3). These regions are selected so that the countries in question have common geographical features (e.g. region 1,

Western coastal Europe with the ocean upwind) or approximately the same level of economic development (e.g. region 4 Northern Europe NO, SE and FI). Fig. 2 displays a map of Europe with the different regions.

Table 3.   Overview of how the stations are grouped in regions for our comparison

Region	Countries/stations	Symbol in the plots
1. Western coastal Europe	Portugal (PT), Spain (ES), Ireland (IE), GB0006R (Lough Navar)	Black plus signs
2. England, Scotland, Wales, English Channel	GB, FR0005R (La Hague)	Cyan circles
3. Continental Europe	Austria (AT), Belgium (BE), Switzerland (CH), Germany (DE), Denmark (DK), France (FR), Netherlands (NL)	Orange asterisks
4. Northern Europe	Norway (NO), Sweden (SE), Finland (FI)	Blue St Andrew crosses
5. Mediterranean	Greece (GR), Italy (IT), Turkey (TR)	Red squares
6. Eastern Europe	Estonia (EE)[a], Hungary (HU), Lithuania (LT), Latvia (LV), Poland (PL), Russia (RU), Slovakia (SK)	Green diamonds

[a]There are other stations in this region, but they do not observe for at least two of the years considered.

3. Results

3.1. Comparison of model results with observations: 1985–1995–2000

Annual mean surface concentration of sulphate is shown in Fig. 3. A few features are worth pointing out. A substantial decrease in sulphate from 1985 to 1995 is found. From 1995 to 2000 sulphate values levelled off. Maximum concentrations are found in Continental Europe and Eastern Europe (1985) for all three sets of emission inventories although Smith et al. (2004) gives lower maximum for 1985 and larger enhanced areas that extends to the east and south than the other two emission inventories.

Before studying the trends we need to establish whether the model is able to reproduce the observed surface concentrations. Figs. 4 and 5 depict a comparison of observed and modelled yearly average concentrations of SO_2 and sulphate (see Table 3

for colour codes). The model both under and overestimates the SO_2 observations for 1985 (i.e. there is a wide spread in the plotted points) while it strongly overestimates SO_2 observations for 1995 and 2000. The EMEP runs show higher correlation coefficients ($r = 0.49$–0.63) for SO_2 than the other two sets, i.e. EMEP overestimates the observations, but in a consistent way. For sulphate the model reproduces well the observations (most stations within 50% deviation). For 1985 there is a larger spread in the modelled/observed values and low correlation coefficients ($r = 0.28$–0.32), regardless of the emission inventory chosen (as seen in SO_2). For 1995 and 2000 the correlation coefficients are between 0.40 and 0.60. The model underestimates sulphate in region 1 (Western coastal Europe) for 1995 and 2000: this will be discussed later.

The differences in the SO_2/sulphate pattern between model and observations need further consideration. There are several reasons for such deviations. Observation sites sample at the ground while model results are taken from the lowermost layer
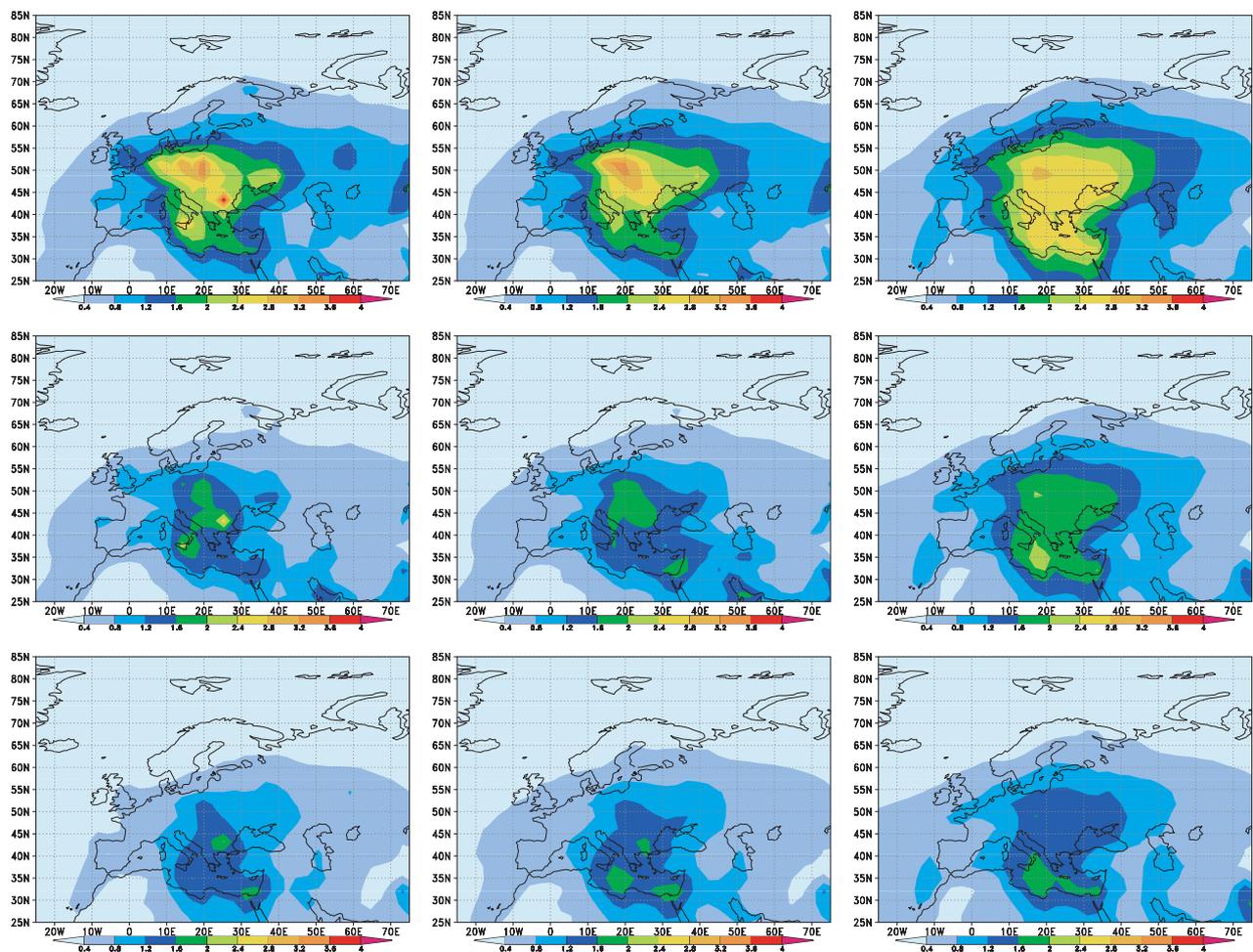
Fig. 3. $SO_4{}^{2-}$ model concentration, annual mean, 1985 (upper row), 1995 (middle), and 2000 (lower row) using EMEP (left-hand column), GEIA/EDGAR/AEROCOM (middle) and Smith et al. (right-hand column) emission inventories. Unit: μg m^{-3}.
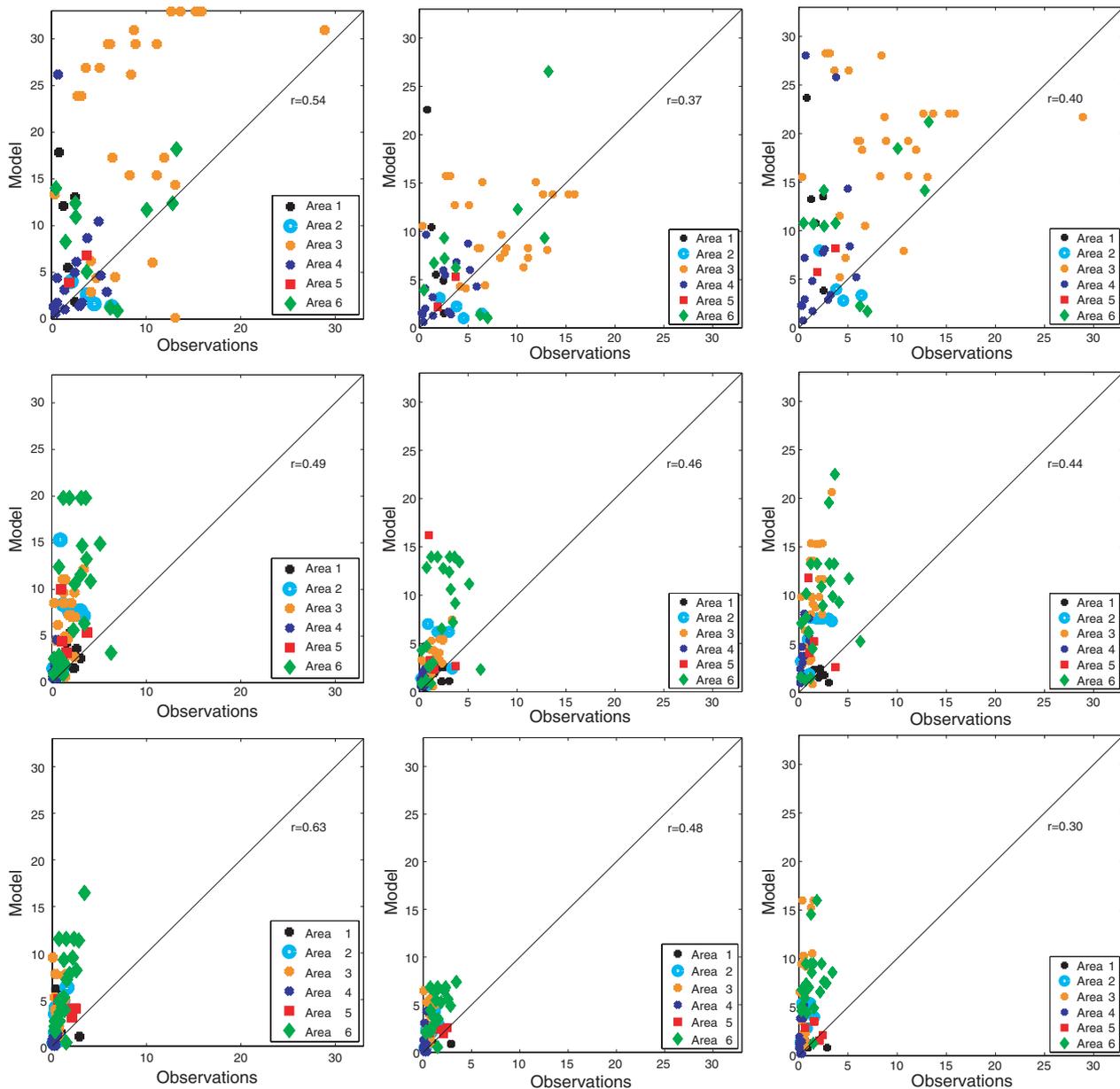
Fig. 4. SO$_2$ model concentration versus observations, annual mean, 1985 (upper row), 1995 (middle) and 2000 (lower row) using EMEP (left-hand column), GEIA/EDGAR/AEROCOM (middle) and Smith et al. (right-hand column) emission inventories. Unit: μg m^{-3}. Annual mean of the observations is elaborated using monthly observations (including stations with at least 7 months of observations). See Table 3 for explanation of colours/symbols. Correlation coefficients are included. Maximum value in plot: 32.9

(20-m thick). Values of SO$_2$ are determined by SO$_2$ emitted at the ground, dry deposition, gas phase oxidation and boundary layer mixing. Sulphate at the ground is either due to gas phase oxidation by OH or due to boundary layer mixing from above of oxidized sulphate as there are no clouds in layer 1 in the model and therefore no aqueous phase oxidation. Like other studies on the sulphur cycle (e.g. Koch et al., 1999; Barth et al., 2000; Berglen et al., 2004) these model runs also show a strong oxi-

dation limitation in wintertime (monthly averages not shown), i.e. low abundance of oxidants give reduced oxidation and hence high SO$_2$ and low sulphate. The annual mean values reported here are influenced by this high SO$_2$/low sulphate values in winter. Chin et al. (2000b) also reported this high SO$_2$/low sulphate pattern and suggest that sea salt in the observation data may partly explain this. Boucher and Pham (2002) overestimate sulphate in Europe, but do not report SO$_2$. There may be several explanations
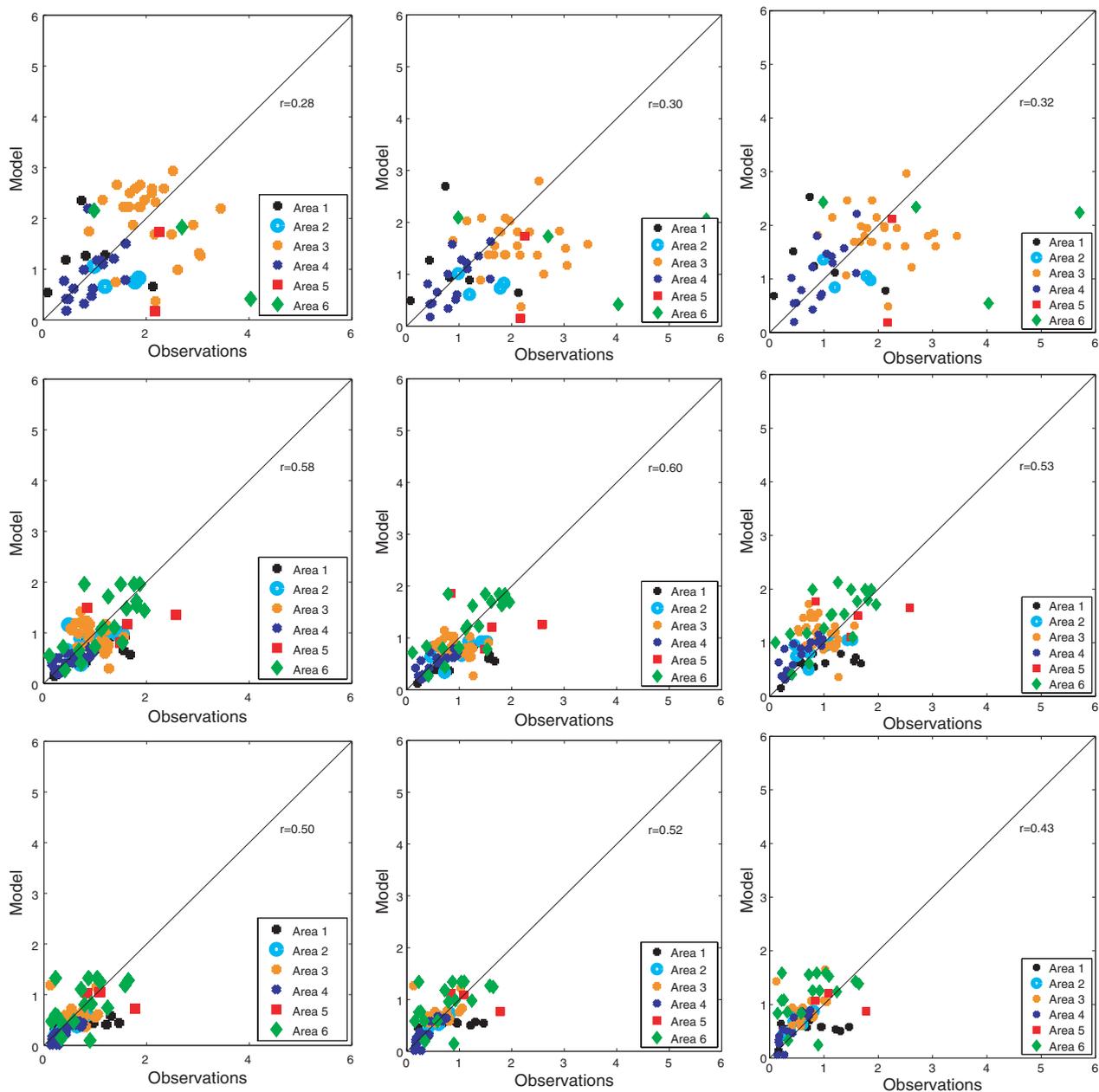
Fig. 5. Same figure, but for SO_4^{2-}. Unit: μg m^{-3}. Maximum value in plot: 5.9.

for the too high SO_2/sulphate distribution and further studies are required.

3.2. Trends in observed concentrations

Figures 4 and 5 (*x*-axis) show a substantial decrease in observed SO_2 and sulphate from 1985 to 1995, while from 1995 to 2000 there was some decrease although somewhat smaller reductions per year in this latter period. Countries that are grouped together

are quite homogeneous with approximately same levels of observed sulphate. As seen in Fig. 3 the highest sulphate levels were observed over Continental Europe (region 3) in 1985, whereafter these regions experienced a considerable decrease. Countries in Eastern Europe (region 6) generally show the highest sulphate concentrations in 1995 and 2000.

Figure 6 shows the trends in sulphate concentrations from 1985 to 1995 (32 stations considered), 1995–2000 (55 stations) and 1985–2000 (22 stations), plotted as percent change in annual
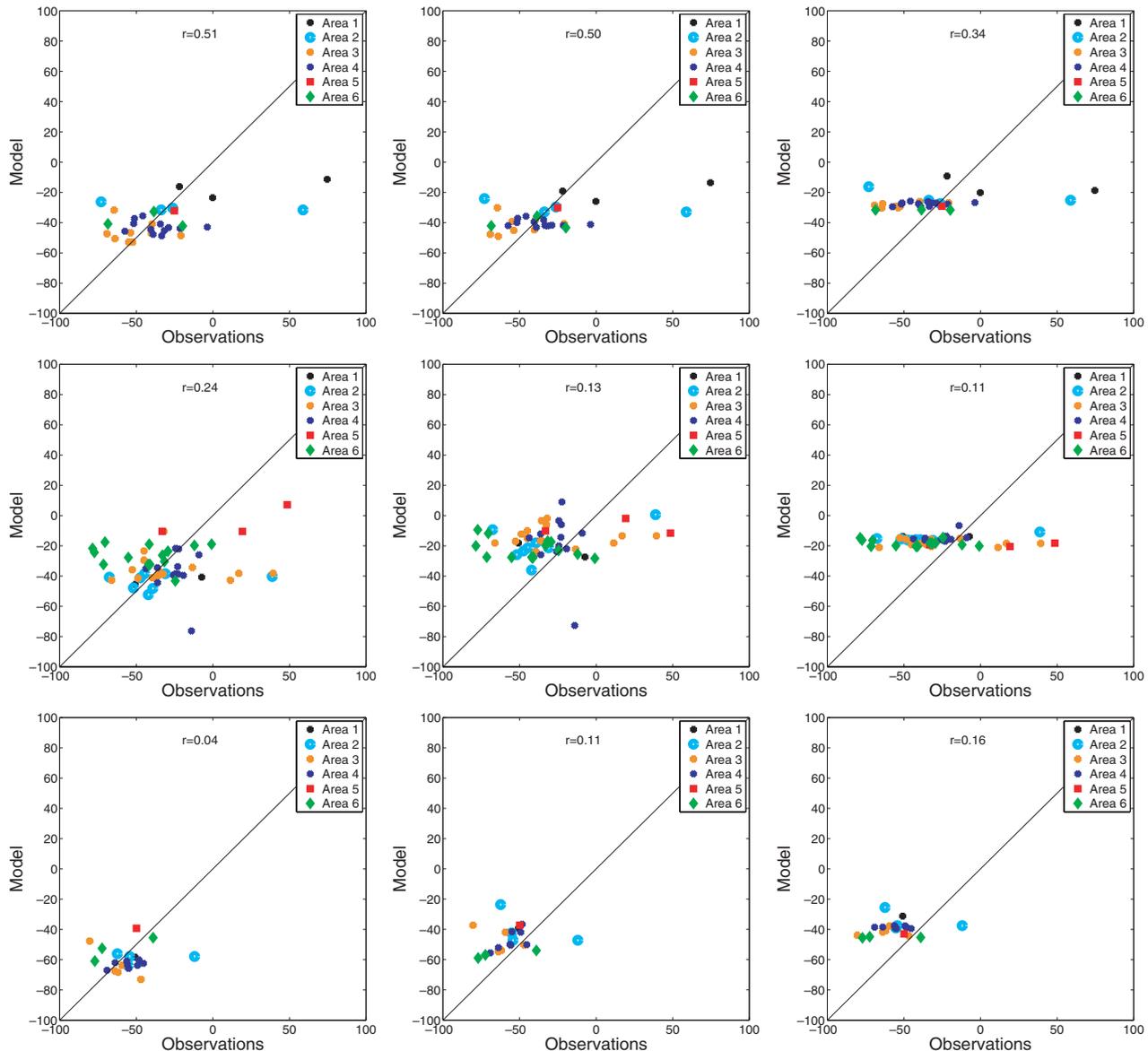
Fig. 6. Trends of sulphate, percent change, 1985–1995 (upper row), 1995–2000 (middle) and 1985–2000 (lower) using EMEP (left-hand column), GEIA/EDGAR/AEROCOM (middle column) and Smith et al. (right-hand column) emission inventories, model results versus observations. Correlation coefficients are included.

means, observations versus model. Concerning the observations we see that from 1985 to 1995 all stations except two (EMEP codes ES0001R and GB0007R) experience a considerable decrease in the observed concentrations. From 1995 to 2000 seven stations report an increase. Here we have plotted the numbers in percent. Plots using absolute numbers (not shown) generally show the same picture, except that for the countries in Western coastal Europe and Northern Europe (regions 1 and 4) the decrease in concentration is small but considerable in percent (20–40%) due to low observed values initially. However, the general analysis of the trends is the same whether we use percent or absolute numbers: The correlation coefficients in the 1985–1995

plots are higher ($r = 0.34$–0.51) than during the other two time periods. But some single points/stations may alter the correlation coefficients considerably, like the two stations mentioned earlier ('out layers').

In Eastern Europe (region 6) only a few stations observed sulphur prior to 1990. New stations were established from mid 1990s and onwards. From 1995 to 2000 sulphur decrease substantially in some parts of Eastern Europe (80%) while other parts show little change in sulphur levels. For the three stations continuously monitoring sulphate over the 1985–2000 period there was a substantial decrease in sulphate levels.

3.3. Modelling the observed sulphate trends

When we compare model results and observations (scatter plot Fig. 6) we note that the observations show a large span in values [from −80 to 80% (1985–1995) and −80 to + 40% (1995–2000)], while the model results show a 20–40% decrease (1985–1995) and between 0 and 40% decrease (1995–2000).

We have listed the percentage change in sulphate per region in Table 4. In all three sets of emissions, the observations decrease more than the estimated concentrations. For example, observations from 1985 to 2000 decrease by 59% while the Em, GEA and Sm inventories estimate 53, 52 and 55% decrease, respectively. Note that observed sulphate in Europe is reduced by more than 50% from 1985 to 2000 for all the regions investigated.

Region 3 (Continental Europe) and region 6 (Eastern Europe) experience the largest decrease over the period; −65 and −63% in observed sulphate concentrations, respectively. The reductions are mostly due to implementation of new abatement technologies and switching of fuel in region 3 and economic recession/transition in region 6, although cleansing technologies have been implemented in Eastern Europe from the middle of the 1990s as well. All the three inventories also estimate a large decrease in emissions in these two regions and hence region 3 and 6 are the regions with largest modelled decrease in most cases. The emissions decrease more than the model results, i.e. there is a certain damping of the signal from the emissions on the model results.

The model is not able to catch the 6% increase in observed sulphate for the Western coastal Europe (region 1) from 1985 to 1995 as the model reports a 16–19% decrease. The GEA set of inventories have a 11% increase in emissions if we look at the three grid boxes with stations only, but 25% decrease if we look at the entire region (numbers not shown). Influence from increasing ship emissions not captured by the emission inventories is one possible explanation for this discrepancy (Endresen et al., 2003; Communication from the Commission to the European Parliament and Council COM 595, 2002). Note that the Smith et al. (2004) inventory from ships increases. See Section 4 for further discussion on ship emissions.

Region 4 (Northern Europe) has a large number of stations observing sulphur. The model compares well with observed trends. To look at percent change in this region may be a bit misleading since the values are low compared to the rest of Europe. A large part of the observed sulphate is transported from sources outside the region, (e.g. Great Britain, overseas, see Klein et al., 2005). Another evidence for long-range transport into the region is that the emission inventories estimate an increase while both observations and model results decrease with reasonable agreement.

Region 5 (Mediterranean) has very few stations to validate our results (1 and 3 stations for the two time periods, respectively) and we should be careful to emphasize on these numbers too much. In addition, sulphur in this region may be highly influenced by local ship traffic.

For the emissions (results not shown) the percent change varies considerably depending on whether we calculate the mean based on the grid boxes containing observation sites only or the entire region, sometimes even the sign of the changes differ. Sulphur

Table 4. Percent change in sulphate (SO$_4{}^{2-}$) observations and model results, 1985–1995 (upper section), 1995–2000 (mid-section) and 1985–2000 (lower section). Results for each region and total. Model results are sampled in grid boxes containing an observation site. See Table 3 and Fig. 2 for definitions of regions

Period	Reg. 1	Reg. 2	Reg. 3	Reg. 4	Reg. 5	Reg. 6	Total
1985–1995							
Obs 1985–1995	6	−29	−53	−36	−25	−55	−44
Model$_{EMEP}$ 1985–1995	−17	−30	−48	−44	−32	−39	−41
Model$_{GEA}$ 1985–1995	−19	−31	−43	−41	−30	−41	−39
Model$_{Smith}$ 1985–1995	−16	−24	−29	−28	−29	−32	−28
Number of stations	3	4	9	12	1	3	32
1995–2000							
Obs 1995–2000	−24	−42	−32	−24	1	−38	−32
Model$_{EMEP}$ 1995—2000	−33	−31	−23	−8	−4	−21	−21
Model$_{GEA}$ 1995–2000	−24	−20	−14	−12	−9	−24	−18
Model$_{Smith}$ 1995–2000	−14	−15	−18	−15	−19	−19	−18
Number of stations	2	8	15	12	3	15	55
1985–2000:							
Obs 1985–2000	−51	−50	−65	−53	−50	−63	−59
Model$_{EMEP}$ 1985–2000	−49	−48	−58	−47	−38	−54	−52
Model$_{GEA}$1985–2000	−39	−42	−49	−46	−37	−57	−49
Model$_{Smith}$ 1985–2000	−31	−35	−42	−39	−43	−45	−41
Number of stations	1	4	5	8	1	3	22

may be transported several hundred kilometres from its source until it is converted a few days later, so sulphate observed at a site in one grid box is influenced by the emission in the adjacent grid boxes/areas upwind. To analyse the wind directions/wind speed and include emissions from these grid boxes would probably give a more consistent picture.

For regions 2 and 3 (1985–1995), 2, 3 and 5 (1995–2000), and 1–4 (1985–2000) and for the total (all three periods) the EMEP inventories give model values close to the observations. Based on these results we therefore conclude that the model runs with the EMEP inventory best reproduce the observed trends. From Fig. 6 we see that the correlation coefficient for EMEP is low ($r = 0.04$ for 1985–2000), but the results are centred around the 1:1 line.

The effects of different inventory construction methods are also apparent in Fig. 6. Modelling results using the Smith et al. (2004) inventory show a much smaller range of variation than the other two inventories. This is due to the construction methodology for this inventory where sources from year-to-year are all scaled together within a region. This method was used because this inventory extends over 150 yr and was intended for long-term modelling efforts. Electric power plant emissions over all

of Western Europe, for example, were scaled together in the girding scheme. The regionally based EMEP inventory contains more spatial variation in emissions from year-to-year. Even using the EMEP inventory, however, the modelled variation is less than that seen in the observations. This could be due to a combination of factors, such as further spatial variability still unaccounted for in the inventories, finite model spatial and temporal resolution, subgrid scale (or timescale) meteorological variability, or other smoothing effects in the model.

To investigate how changes in sulphur emissions have changed the loss processes, we made a table showing the sulphur emissions and loss pathways (Table 5). There is a certain long-range transport into Europe from areas up winds, mainly from North America. With a sulphur lifetime of the order of a few days, some sulphur emitted overseas will reach Europe and will be deposited. For example, Tarrasón et al. (2005) estimate that ∼10% of sulphur deposited in Europe originates from sources outside Europe. Net export of sulphur out of Europe = emissions − deposition + import. In Table 5 the emissions in Europe are larger than loss for all inventories/years. Hence there is a net export of sulphur out of Europe and the difference between

Table 5. Change in total emissions and loss of sulphur in Europe for the three emission inventories used in this study. For loss of SO_2, the mass and fraction that is deposited (wet and dry deposition) and oxidized to sulphate is reported. Percent changes in emissions and loss from 1985 to 1995 and 1995 to 2000 are also listed. Unit for mass is Tg(S) yr^{-1}. See Fig. 2 for the area defined as Europe

	Emis. S	Loss S	Dep. SO_2		Ox. SO_2	Dep. SO_4^{2-}
EMEP						
1985	25.22	19.46	7.44		13.43	12.02
Dep. SO_2/ox. to SO_4^{2-}				36%/64%		
Change 1985–1995%	−44	−41	−46		−38	−38
1995	14.11	11.47	3.99		8.27	7.48
Dep. SO_2/ox. to SO_4^{2-}				33%/67%		
Change 1995–2000	−16	−12	−17		−4	−9
2000	11.80	10.13	3.31		7.91	6.82
Dep. SO_2/ox. to SO_4^{2-}				29%/71%		
GEA						
1985	25.75	20.39	6.82		15.04	13.57
Dep. SO_2/ox. to SO_4^{2-}				31%/69%		
Change 1985–1995	−41	−39	−45		−36	−36
1995	15.10	12.48	3.76		9.62	8.72
Dep. SO_2/ox. to SO_4^{2-}				28%/72%		
Change 1995–2000	−18	−14	−26		−5	−9
2000	12.41	10.70	2.78		9.17	7.92
Dep. SO_2/ox. to SO_4^{2-}				23%/77%		
Smith et al.						
1985	34.53	27.20	9.84		20.70	17.36
Dep. SO_2/ox. to SO_4^{2-}				32%/68%		
Change 1985–1995	−42	−37	−45		−33	−32
1995	19.89	17.17	5.38		13.89	11.79
Dep. SO_2/ox. to SO_4^{2-}				28%/72%		
Change 1995–2000	−22	−18	−22		−17	−16
2000	15.47	14.05	4.17		11.53	9.88
Dep. SO_2/ox. to SO_4^{2-}				27%/73%		

emissions and deposition (emissions − deposition) will then be a lower limit for the net export of sulphur. Concerning our study, the importance of long-range transport is limited though; first the amount of sulphur emitted in Europe is much larger than the amount brought to Europe from elsewhere. And second the North American sources show approximately the same decreasing trend as European sources. Hence these upwind sources will barely alter our calculated trends found in Europe. This net export out of Europe is decreasing, from over 5 Tg(S) in 1985 to ∼1.5 Tg(S) in 2000 (long-range transport into Europe not accounted for). Otherwise we see that both emissions and loss decrease from 1985 to 2000 although the reductions is strongest from 1985 to 1995 (both over the period and per year). The total loss decreases while the percentage oxidized to sulphate increases. Hence the effect from emission reduction upon sulphate is damped. This is in agreement with results found in Berglen et al. (2004). If we look at the oxidation of SO_2 and deposition of sulphate we see an interesting detail. For most cases the percent change of these two loss pathways is the same, except for EMEP and GEA for 1995–2000 (−4% vs. −9% and −5% vs. −9%, respectively). We have two explanations for this discrepancy, it is either due to sulphate transported from elsewhere affecting our calculated numbers. Or more likely it is due to the fact that the fraction Dep. SO_4^{2-}/Ox. SO_2 is lower in the year 2000 than in 1985 and 1995. Hence relatively less sulphate is deposited, probably transported out of the European region. In our Berglen et al. (2004) study we also found that the variation in the fraction of SO_2 oxidized to sulphate is most sensitive to changes in sulphur emissions, and to a lesser extent to changes in oxidants and emissions of oxidants. In any case atmospheric chemistry and the sulphur cycle is a complex non-linear system that will change with changing emissions.

3.4. Robustness and sensitivity of the results

All these model runs were performed with the same meteorology representing the year 2000. To see how sensitive these model runs are with regard to changes in meteorology we performed model runs with the EMEP 1985/1995/2000 inventories with meteorology representing the year 1997 (same spin-up procedure and otherwise identical to the EmXX runs listed in Table 1). Results for the 2000 and the 1997 runs are seen in Fig. 7 (percent change). For some stations the meteorological conditions may play a role, but the dynamics do not affect the overall picture. The correlation coefficients do not vary significantly.

To investigate to what extent our results are resolution dependent we have performed a test running our model in T21 (5.625° × 5.625°), T42 (2.8° × 2.8°) and 1° × 1° horizontal resolutions for the months of January and July (results not shown). Vertical resolution was 40 layers, and the model was run with full tropospheric chemistry with sulphur cycle included (as described in Section 2). All model runs were identical except horizontal resolution; same meteorology and same emission fields were used

for all three resolutions. A few general features must be pointed out. Maximum concentrations for a certain gas increased with finer resolution. This is due to a more detailed emission pattern in the 1° × 1° resolution with high emissions in some specific spots, whereas these high local emissions will be smoothed out in a coarse resolution. Concerning total mass or lifetime of a specific gas there was a considerable difference between T21 on one side and T42/1° × 1° on the other side, i.e. that the T42 and 1° × 1° were quite similar, concerning, e.g. total mass and total lifetime of the most important species, and also concerning total wind generated DMS emissions, and SO_2 loss processes. We therefore claim that T42 is suitable to capture the basic features of the sulphur cycle. To increase model resolution from T21 as used in Berglen et al. (2004) to T42 in this study improved model performance substantially (verified by comparison with observations). To use even finer grid (1° × 1°) will to a certain extent improve the model, but the major advancement was obtained by switching from T21 to T42.

4. Discussion and conclusions

As seen in both the observations and from the emission inventories there has been a strong reduction of sulphate in Europe during the last two decades. This is partly due to implementation of abatement technologies in Western Europe and partly to economic recession in Eastern Europe. However, the decrease of European sulphate has levelled off during the last few years and sulphate concentrations are reported to have even increased slightly in some regions.

The aim of this study was to investigate the negative trend in sulphate concentrations observed over Europe during the last two decades as a result of reduced emissions. The trend has been modelled based on different published emission inventories for three different years (1985, 1995 and 2000) using the Oslo CTM2 model. The CTM2 model results were compared with observations from the EMEP network. SO_2 and sulphate were investigated, although trends of sulphate were our main concern. To facilitate the comparison the countries were grouped into six different geographical/economic regions.

While the model agrees reasonably well for sulphate for all three sets of inventories, it tends to overestimate SO_2. This is a pattern seen in many previous studies of the sulphur cycle. An oxidation limitation leading to high SO_2/sulphate ratio in winter due to low abundance of oxidants was identified as a possible source of discrepancy, see Section 3.1 for a discussion on this matter.

The model is able to catch the trends in observed sulphate concentration, although the model shows a slightly smaller decrease than the observations. Observations from the 22 stations monitoring sulphate from 1985 to 2000 show a 59% decrease throughout Europe for 1985–2000 while we model a 52, 49 and 41% decrease using the EMEP, GEA and Smith et al. (2004) inventories, respectively. The two regions Continental Europe and
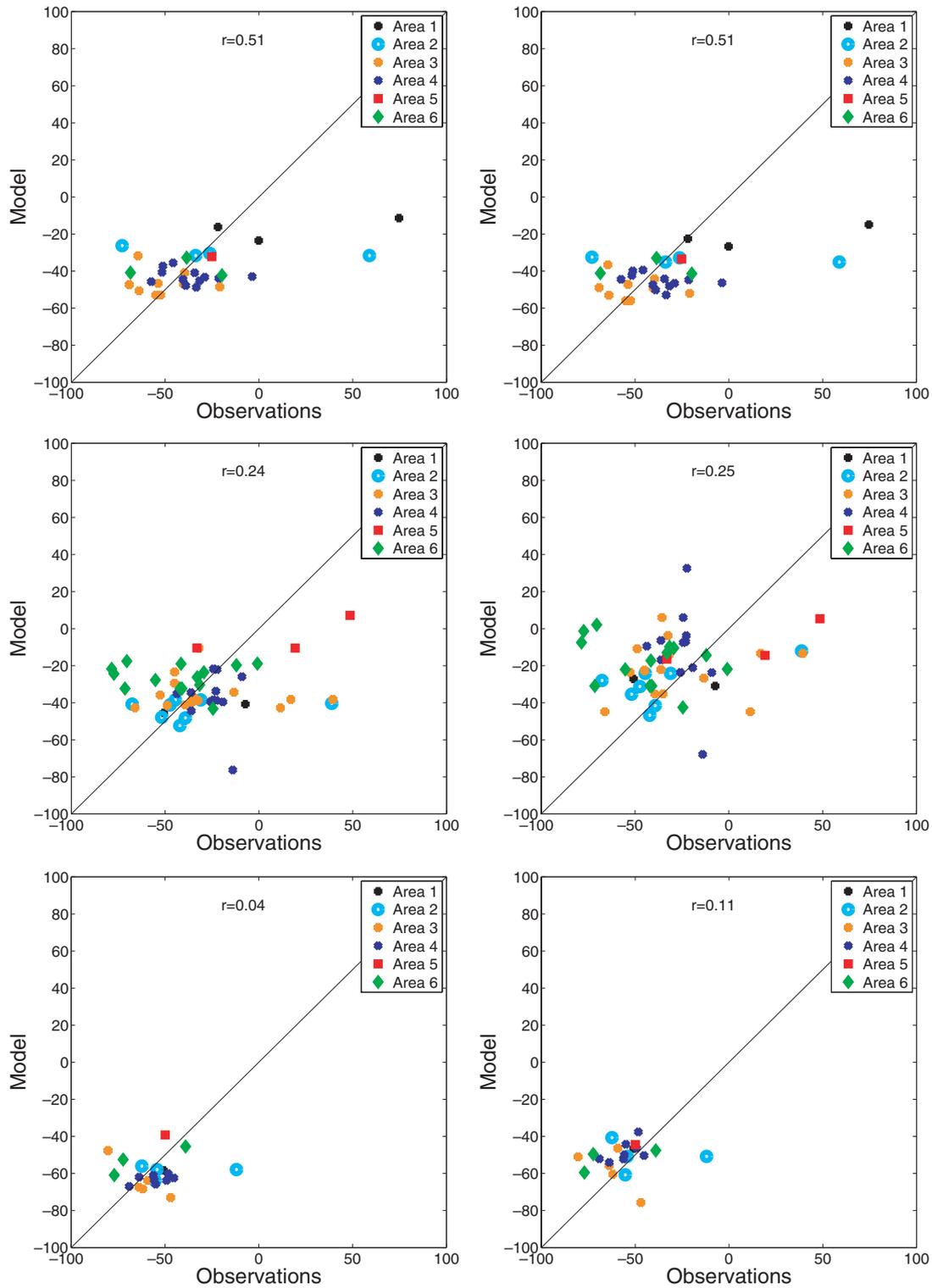
Fig. 7. Trends of sulphate, percent change, 1985–1995 (upper row), 1995–2000 (middle) and 1985–2000 (lower) using EMEP emission inventories, 2000 meteorology (left-hand column), 1997 meteorology (right-hand column), model results versus observations. Correlation coefficients are included.
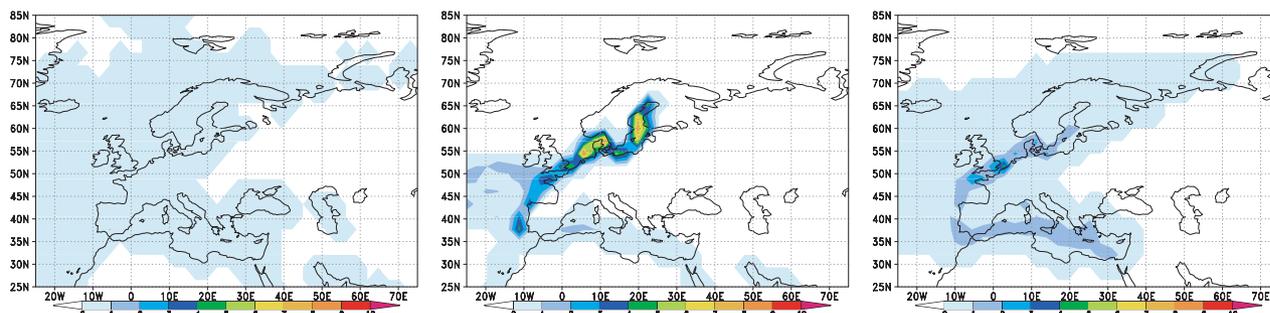
Fig. 8. Ship emissions for the year 2000, AMVER (left), AEROCOM (middle) and EMEP (right) inventories. Unit: 10^{10} molec. m^{-2} s^{-1}.

Eastern Europe experienced the largest decrease over the period we studied; 65 and 63% decrease in observations, respectively, and between 42 and 58% decrease in model estimates.

Observed sulphate increases in Western coastal Europe from 1985 to 1995, this is not captured by our model simulations. Although the number of stations is limited (three) we can see a slight increase. A possible explanation for this discrepancy is that emissions from ships have increased substantially during the last few decades, while other anthropogenic emissions have decreased, and that present emission inventories underestimate this increase. Ship emissions constitute a large part of the total emissions in costal areas, and hence a large part of the observed sulphate as there is only minor emission upwind. In Fig. 8 we have shown three examples of ship emission inventories for year 2000 (AMVER, AEROCOM and EMEP). These inventories differ significantly, and will give very different results when applied in the model. The AEROCOM inventory has much larger emissions in Europe, both along the coast of Europe and in the North Sea and in the Baltic Sea. To obtain historical emissions one method is to scale emissions backwards, assuming an annual increase varying typically between 1.5 and 2.5% (AMVER is scaled by 1.6% in the Em and GEA model runs in this study). This method does not take into account that different types of ships have different historic growth rate in emissions. Hence the emission increase, or in some rare cases decrease, will vary both in time and space. Further detailed studies of ship emissions and its impact are clearly needed. Ship emissions are likely to increase in the future and will get increasingly important as ships also affect sulphur levels onshore.

Model runs using meteorological input data for 1997 instead of 2000 show that the dynamics influence on our results is only minor. Changes in sulphur emissions during the period are found to be more important than changes in oxidation processes for the observed decrease in sulphur compounds.

The direct aerosol effect of sulphate is estimated to be as strong as -3 W m^{-2} in certain European regions for the pre-industrial time to 1985 (Myhre et al., 2004). A significant but more uncertain indirect effect for sulphate over Europe for the same time period is also simulated (Lohmann and Feichter, 2005). A reduction in the sulphate over Europe may thus have a warming effect.

Here we see an example of how air pollution policy decisions may impact the climate.

Our overall conclusion is that we are able to model the recent decrease in sulphur reasonably well. Of the three sets of inventories used in this study the model results using the EMEP emission inventory best reproduce the trends in observations. The future sulphate levels and trends in Europe will depend on socioeconomic factors, technology and political decisions. Science and research have proven to be an important factor in sulphate abatement in the past and should certainly be an important contributor in the future.

5. Acknowledgments

References

Amann, M., Bertok, I., Cofala, J., Gyafas, F., Heyes, C. and co-authors. 2005. Baseline Scenarios for the Clean Air for Europe (CAFÉ) Programme, Final Report, Contract No B4-3040/2002/340248/MAR/C1.

Barth, M. C., Rasch, P. J., Kiehl, J. T., Benkovitz, C. M. and Schwartz, S. E. 2000. Sulfur chemistry in the National Center for Atmospheric Research Community Climate Model: cescription, evaluation, features, and sensitivity to aqueous chemistry. *J. Geophys. Res.* **105**, 1387–1415.

Benkovitz, C. M., Scholtz, M. T., Pacyna, J., Tarrasón, L., Dignon, J. and co-authors. 1996. Global gridded inventories of anthropogenic emissions of sulfur and nitrogen. *J. Geophys. Res.* **101**, 29239–29253.

Berglen, T. F., Berntsen, T. K., Isaksen, I. S. A., Sundet, J. K. 2004. A global model of the coupled sulfur/oxidant chemistry in the troposphere: the sulfur cycle. *J. Geophys. Res.* **109**, D19, D19310, doi:10.1029/2003JD003948

Boucher, O. and Pham, M. 2002. History of sulfate aerosol radiative forcings. *G. Res. Lett.* **29**(9), 1308, doi:10.1029/2001GL014048.

Chin, M., Rood, R. B., Lin, S.-J., Müller, J.-F. and Thompson, A. M. 2000a. Atmospheric sulfur cycle simulated in the global model

GOCART: model description and global properties. *J. Geophys. Res.* **105**(D20), 24671–24687.

Chin, M., Savoie, D. L., Huebert, B. J., Bandy, A. R., Thornton, D. C. and co-authors. 2000b. Atmospheric sulfur cycle simulated in the global model GOCART: comparison with field observations and regional budgets. *J. Geophys. Res.* **105**(D20), 24689–24712.

Communication from the Commission to the European Parliament and Council, A European Union strategy to reduce atmospheric emissions from seagoing ships, COM(2002) 595 final, Bruxelles 20.11.2002. See also: http://europa.eu.int/comm/environment/air/background.htm

Dentener, F., Kinne, S., Bond, T., Boucher, O., Cofala, J. and co-authors. 2006. Emissions of primary aerosol and precursor gases in the years 2000 and 1750 prescibed data-sets for AeroCom. *Atmos. Chem. Phys.* **6**, 4321–4344.

Endresen, Ø., Sørgård, E., Sundet, J. K., Dalsøren, S. B., Isaksen, I. S. A. and co-authors. 2003. Emission from international sea transportation and environmental impact. *J. Geophys. Res.* **108**(D17), 4560, doi:10.1029/2002JD002898.

Fricke, W. and Beilke, S. 1992. Indications for changing deposition patterns in Central-Europe. *Enviro. Pollut.* **75**(2), 121–127.

Grennfelt, P. and Hov, Ø. 2005. Regional air pollution at a turning point. *Ambio* **34**(1), 2–10.

Hesstvedt, E., Hov, Ø. and Isaksen, I. S. A. 1978. Quasi steady-state approximation in air pollution modelling: Comparison of two numerical schemes for oxidant prediction. *Int. J. Chem. Kinet.* **10**, 971–978.

Hjellbrekke, A.-G. 2005. Data Report 2003, Acidifying and eutrophying compounds, EMEP/CCC-Report 3/2005 (http://www.nilu.no/projects/ccc/reports.html).

Holtslag, A. A. M., DeBruijn, E. I. F. and Pan, H. L. 1990. A high resolution air mass transformation model for short-range weather forecasting, *Mon. Wea. Rev.*, **118**, 1561–1575.

Iversen, T. and Seland, Ø. 2002. A scheme for process-tagged SO4 and BC aerosols in NCAR CCM3: validation and sensitivity to cloud processes, *J. Geophys. Res.* **107**(D24), 4751, doi:10.1029/2001JD000885.

Kettle, A. J. and Andreae, M. O. 2000. Flux of dimethylsulfide from the oceans: A comparison of updated data seas and flux models. *J. Geophys. Res.* **105**(D22), 26793–26808.

Kettle, A. J., Andreae, M. O., Amouroux, D., Andreae, T. W., Bates, T. S. and co-authors. 1999. A global database of sea surface dimethylsulfide (DMS) measurements and a procedure to predict sea surface DMS as a function of latitude, longitude and month. *Global BioGeochem. Cycles* **13**, 399–444.

Klein, H., Wind, P. and van Loon, M. 2005. Transboundary air pollution by main pollutants (S, N, O₃) and PM. MSC-W Data Note 1/2005, ISSN 0804–2446.

Koch, D., Jacob, D. J., Tegen, I., Rind, D. and Chin, M. 1999. Tropospheric sulfur simulation and sulfate direct radiative forcing in the Goddard Institute for Space Studies general circulation model. *J. Geophys. Res.* **104**, 23799–23822.

Lelieveld, J., Berresheim, H., Borrmann, S., Crutzen, P. J., Dentener, F. J. and co-authors. 2002. Global air pollution crossroads over the Mediterranean, *Science* **298,** 794–799.

Lohmann, U. and Feichter, J. 2005. Global indirect aerosol effects: a review. *Atmos. Chem.Phys.* **5,** 715–737.

Lövblad, G., Tarrasón, L., Tørseth, K. and Dutchak, S., (eds), 2004. EMEP Assessment, Part 1, European Perspective. EMEP-MSC-W, Oslo. http://www.emep.int/index_assessment.html

Myhre, G., Stordal, F., Berglen, T. F., Sundet, J. K. and Isaksen, I. S. A. 2004. Uncertainties in the radiative forcing due to sulfate aerosols. *J. Atmos. Sci.* **61,** 485–498.

Mylona, S. 1996. Sulphur dioxide emissions in Europe 1880–1991 and their effect on sulphur concentrations and depositions. *Tellus B* **48**(5), 662–689., doi:10.1034/j.1600-0889.1996.t01-2-00005.x

Nightingale, P. D., Malin, G., Law, C. S., Watson, A. J., Liss, P. S. and co-authors. 2000. In situ evaluation of air-sea gas exchange parameterizations using novel conservative and volatile tracers. *Global BioGeochem. Cycles* **14,** 373–387.

Olivier, J. G. J. and Berdowski, J. J. M. 2001. Global emissions sources and sinks. In: *The Climate System* (eds.J. Berdowski, R. Guicherit and B. J. Heij). A.A. Balkema Publishers/Swets & Zeitlinger Publishers, Lisse, The Netherlands, 33–78.

Prather, M. J. 1986. Numerical advection by conservation of 2nd order moments. *J. Geophys. Res.* **91**(D6), 6671–6681.

Rasch, P. J., Barth, M. C., Kiehl, J. T., Schwartz, S. E. and Benkovitz, C. M. 2000. A description of the global sulfur cycle and its controlling processes in the National Center for Atmospheric Research Community Climate Model, Version 3. *J. Geophys. Res.* **105**(D1), 1367–1385.

Rodhe, H. and Isaksen, I. S. A. 1980. Global distribution of sulfur-compounds in the troposphere estimated in a height-latitude transport model. *J. Geophys. Res* **85**(NC12), 7401–7409 1980.

Rotstayn, L. D. and Lohmann, U. 2002. Simulation of the tropospheric sulfur cycle in a global model with a physically based cloud scheme, *J. Geophys. Res.* **107**(D21), 4592, doi:10.1029/2002JD002128.

Smith, S. J., Andres, R., Conception, E. and Lurz, J. 2004. Historical Sulfur Dioxide Emissions 1850–2000: Methods and Results. PNNL Research Report, PNNL-14537.

Spiro, P. A., Jacob, D. J. and Logan, J. A. 1992. Global inventory of sulfur emissions with $1° \times 1°$ resolution. *J. Geophys. Res.* **97**, 6023–6036.

Tarrasón, L., Benedictow, A., Fagerli, H., Jonson, J. E., Klein, H., *et al.* 2005. Transboundary acidification, eutrophication and ground level ozone in Europe in 2003. EMEP Report 1/2005, ISSN 0806-4520.

Vestreng, V., Adams, M. and Goodwin, J. 2004. Inventory Review 2004, Emission Data reported to CLRTAP and under the NEC Directive, EMEP/EEA Joint Review Report, EMEP/MSC-W Note 1/2004. ISSN 0804-2446.