

European air pollution emission trends – review, validation and application

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*To Vemund
with the best hopes for a
sustainable future*

SPØR IGJEN

Tallrekken ler av oss
og vil forklare alt.
Den har kjeve av jern og tenner
som det klirrer i.

Vi spør og vi spør
og tallene svarer
men ikke om fiolinene
eller om lykken mellom to armer.
Da hoster det på skjermen:
– uklart spørsmål.
Spør igjen.

ASK AGAIN

The row of numbers laughs at us
and wants to explain everything.
It has jaws of iron and clattering
teeth.

We ask and ask
and the numbers answer,
but not of violins
or the joy of being embraced.
The screen will cough
– unclear question.
Ask again.

Rolf Jacobsen (1907-1994), Nattåpent (1985). Translated by Olav Grinde

List of publications

This thesis consists of the following five papers :

- I. Vestreng, V., Myhre, G., Fagerli, H., Reis, S., and Tarrasón, L.: Twenty-five years of continuous sulphur dioxide emission reduction in Europe, *Atmos. Chem. Phys.*, 7, 3663-3681, 2007.
- II. Vestreng, V., Ntziachristos, L., Semb, A., Reis, S., Isaksen, I. S. A., and Tarrasón, L.: Evolution of NO_x emissions in Europe with focus on road transport control measures. *Atmos. Chem. Phys. Discuss.*, 8, 10697-10747, 2008.
- III. Berglen, T. F., Myhre, G., Isaksen, I.S.A., Vestreng, V., and Smith, S. J.: Sulphate trends in Europe: Are we able to model the recent observed decrease? *Tellus*, 59B (4), 773-786, 2007.
- IV. Fagerli, H., Legrand, M., Preunkert, S., Vestreng, V., Simpson, D., and Cerqueira, M.: Modeling historical long-term trends of sulfate, ammonium, and elemental carbon over Europe: A comparison with ice core records in the Alps. *J. Geophys. Res.*, 112, D23S13, doi:10.1029/2006JD008040, 2007.
- V. Marmer, E., Langmann, B., Fagerli, H., and Vestreng, V.: Direct shortwave radiative forcing of sulphate aerosol over Europe from 1900 to 2000. *J. Geophys. Res.*, 112, D23S17, doi:10.1029/2006JD008037, 2007.

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It was not evident that my project; to re-educate from a Master of Science at the Norwegian polytechnic university (NTH) and senior petroleum geophysicist via a doctor scient study to an air pollution emission expert, would turn out successfully. My supervisor, Ivar S. A. Isaksen, was however both enthusiastic and supportive when I turned up in his office as pregnant as few, and older than most PhD students and told him that I had looked his research up in the University of Oslo (UIO) booklet “We are doing research”, and asked to be one of his students. Ivar deserves many thanks for being so open minded and helpful from the very beginning of this study. Along the way from my re-entry at the UIO in 1997, maternity leave in France, several applications with little success to get a scholarship, and to a permanent position at the Norwegian Meteorological Institute (met.no) in 2000, the considerate help given to me by Kristin Rypdal is heartily acknowledged. At met.no I was taken under the fast flying wings of Leonor Tarrasón. She also volunteered to be my co-supervisor. I am most grateful for her dedicated guidance and to met.no where this work has been performed. Motherhood and full time work at met.no turned out to be more than enough the following years. Hence it was not until 2006, when Gunnar Myhre offered me his support in publishing the European sulphur emission trends in the peer reviewed literature that the realization of my project put on speed.

My beloved son, Vemund, was backing me up by accepting the even longer working hours and shifting spirits. Reading loud Harry Potter and the Deadly Hollow gave us the required breaks and fun in front of the fire through the dark winter evenings. The music and musicians in Oslo Chamber Orchestra, outdoor activities and Friskis&Svettis, all helped me to keep my shoulders down. Finally, I am in great debt to my best friend, Mathilde Skoie, for her active participation in most aspects of my life, and for weekly correspondent letters from Bergen, distracting me with other modes of intellectual and human discussions.

Content

List of publications

Acknowledgements

| | | |
|-----|---|----|
| 1 | Background and motivation | 1 |
| 2 | Inventory requirements..... | 5 |
| 2.1 | Emission sources of sulphur and nitrogen oxides | 5 |
| 2.2 | Emission estimation approaches..... | 7 |
| 2.3 | Inventory requirements for model applications..... | 11 |
| 2.4 | Data quality | 13 |
| 3 | Results | 16 |
| 3.1 | European emission trends..... | 16 |
| 3.2 | Top-down validation of emission trends | 20 |
| 3.3 | Impact of emission trends..... | 21 |
| 4 | Future perspectives | 22 |
| | References | 26 |
| | Appendix I: Summary of papers..... | 30 |

1 Background and motivation

Værre tider; værre syner
gennem fremtidnatten lyner!
Brittens kvalme stenkulsky
sænker sort seg over landet,
smudser alt det friske grønne,
kvæler alle spirer skønne,
stryger lavt, med giftstof blandet,
stjæler sol og dag fra engen,
drysser ned, som askeregnen
over oldtids dømte by.

Yet worse times; worse visions, frightening
pierce the future's night with lightning!
Britain's coal-clouds spread their gloom
on our land, foul, black and legion,
smudging fresh green vegetation,
spreading vile contamination
on the fair shoots where is splashes,
stealing daylight from our region,
drizzling down as did the ashes,
once that ancient city's doom.

Henrik Ibsen (1828-1906), Brand (1866), 5. Act. Translated by John Northam

This thesis deals with long-term trends in European emissions of sulphur and nitrogen oxides. Although the main focus is on the last 25 years, the history of air pollution dates back several centuries and evidence of pollution can even be found in blackened lung tissues in Egyptian mummies (Walker et al., 1987).

In the nineteenth century, Britain was the largest contributor to air pollution in Europe. Nearly 150 million tonnes coal was burnt annually, accompanied by more than two million tonnes of sulphur emissions (Mylona, 1996). Early records of air pollution are particularly well documented in Britain. In his book *The big smoke* Peter Brimblecombe (1987) describes the British historical records of air pollution. Prior to urbanization, indoor air pollution in badly ventilated huts was the main problem. Early evidence of air pollution in Europe is skulls with incidence of sinusitis. These skulls date back to early medieval times, i.e. around 1066 and the Norman (or Viking) conquest of England. The introduction of chimneys, population growth, fuel wood shortage and thus a consequential increase in residential consumption of coal, caused air pollution to become also an outdoor urban problem. The sulphurous smell suggested to medieval minds that the air was unhealthy and the first attempt to control the use of coal was made in London already in 1306. In the 17th century, the coal consumption in the domestic sector increased rapidly, and concern about the influence of air pollution on human health grew. Not only humans, but also buildings and vegetation suffered from the polluted air. At the same time, the population adapted to

the foul air by wearing off-white instead of white clothes, and painted their houses in dull colours.

In the early days of the industrial revolution (late 18th and early 19th century), a fuel switch from coal to less polluting wood was no longer an option due to lack of trees in the vicinity of London. According to Brimblecombe (1987), cleaner coals (anthracite), improved stove design and different combustion processes were investigated. Public concern about air pollution rose, while industrial interests opposed legislation. In 1853, the Smoke Nuisance Abatement Act, which regulated emissions from factories, was adopted. However, since a monitoring network was not operational at the time, possible improvement in the air quality could not be properly quantified. The frequency of foggy days in London increased in the 19th century, and the general feeling was that the amount of air pollution was growing, counter to the expectations after the Smoke Nuisance Abatement Act (Brimblecombe, 1987). The question therefore arose whether there were sources unaccounted for. Arguments for and against air pollution legislation continued, the public movement grew, and bills advocating abatement were issued and rejected. Then, in 1952, London was hit by a smog (fog intensified by smoke) episode, which apparently speeded up the process of policy making. This particular air pollution event, the Great Smog, was triggered by the meteorological conditions and lasted for five days. During the Great Smog, concentrations of smoke and sulphur dioxide increased dramatically. Visibility turned to near zero, transport stalled, and thousands of people died (e.g. <http://www.metoffice.gov.uk>). In 1965, the Clean Air Act, covering both industrial and domestic sources, but leaving out e.g. sulphur emission control, was enacted.

Early indications of the long-range transport character of air pollution are also recorded in Britain. According to Brimblecombe (1987), the 15th century belief that British air pollution ruined French vine crops, is not likely to be correct, although soot could have been carried that far. Much better documented are the episodes of sulphur smelling “black rain” in Scotland in 1862 and 1863 reported by Reverend James Rust, who discounted local sources (Brimblecombe et al., 1986). Reports on soot coloured snow in the southern part of Norway, were linked to the air pollution from Britain already in 1881 (Brøgger, 1881 in Fløisand eds., 2002). Almost a century later, in the 1960s, the decline in the amount of salmon in Norwegian lakes was tentatively coupled to acidification in the precipitation. This is an interesting case in the history of air pollution as it is an example of

the relationship between research and politics, as well as an important step towards international collaboration in order to abate the effects of air pollution. The political interest in the problem increased when it was proposed that also forests could suffer from acid rain. A comprehensive research project, “The effect of acid rain on forest and fish” (SNSF) was launched in 1972, the year Norway got a Ministry of Environment. However, the programme did not only receive praise. A long lasting debate on both the scientific validity of and the political influence on, the programme took place. The main actors were the Minister of Environment, Gro Harlem Brundtland, and Professor of geology at the University of Oslo, Ivan Rosenquist. Rosenquist did not take part in the SNSF research project, but wrote a book where he claimed that the declared fish death was related to the amount of acids already stored in the soils and due to changes in agricultural practice, rather than to acid rain (Rosenquist, 1977). Rosenquist’s scepticism to the results of the ongoing research and his criticism of Norwegian policies received considerable attention. The majority of the research community however found the relation between the effects and acid rain convincing. Consequently, the Norwegian Government decided in the early 1970s to co-operate more closely with international organizations in order to reduce emissions (Fløisand eds., 2002). This turned out to be a wise decision, as such co-operation, under the United Nations (UN) and the Organization for Economic Co-operation and Development (OECD), has proved particularly successful to abate air pollution and its effects.

Initial steps towards international co-operation in the field of air pollution abatement were taken at a UN *Conference on the Human Environment* in Stockholm in 1972. The same year, the OECD initiated an international programme to address Long-range Transboundary Air Pollution (LRTAP). This programme was later located under the UNECE (United Nations Economic Commission for Europe) to form the Convention on LRTAP in 1979. The Convention was formed under the UN to profit from Russian participation in the work to abate air pollution. Russian involvement was a result of i.a. early “east-west” converses for co-operation to solve environmental problems which took place in a meeting held in Oslo during the “cold war” in 1974. The decision from this meeting to collaborate was later referred to in the Final Act of the Conference on Security and Cooperation in Europe (the Helsinki Final Act) which the Russians signed in 1975. This is an excellent example of how strained political relations and environmental concerns can be combined to create a win-win situation. With its eight protocols in force (UNECE,

2004) and a European wide monitoring and modelling network, the Convention has demonstrated its significance as an instrument for the development of long-term strategies for air pollution control. The initial focus of the protocols was in sulphur and nitrogen compounds aiming to control the impact of acidification and eutrophication. The first 1985 Sulphur Protocol was followed by the 1988 Nitrogen Protocol, and subsequent UN Protocols and EU Directives are in force to abate sulphur and nitrogen oxides (SO_x and NO_x). Later protocols extended to other components, in particular volatile organic compounds (VOC), ammonia (NH_3), heavy metals (HM) and persistent organic compounds (POP).

This thesis deals with European emission trends in both a long-term perspective (in the order of decades) and in a historical perspective (covering over a century). High quality emission inventories and trends are important mainly for three reasons: (1) political agreements and decisions to abate adverse effects of air pollution are based on emission data, (2) emission inventories are essential as input to model studies which aid to determine the state of our environment and climate, and (3) monitoring of compliance with agreed national and international emission reduction targets rely on reported, quality assured emission data. Sulphur and nitrogen oxides were the first pollutants subject to policy control in Europe, due to the early awareness of acidification. The largest emission reductions have consequently been obtained for sulphur and nitrogen oxides. Further, emission estimates of these air pollutants are shown to be more reliable than their counterparts. Thus, relatively firm conclusions can be based on the inventories of sulphur and NO_x ($\text{NO}_x = \text{NO} + \text{NO}_2$). These are the main reasons why it is particularly interesting to focus on sulphur and nitrogen oxides emission trends. The trends in SO_x and NO_x considered in this thesis do not only cover the time from the first European wide legislation, but also go back to pre-industrial times. Knowledge about the historical evolution of emissions is essential in order to assess the state of our environment. This is because air pollution accumulates in soils and waters over time. Dynamic modelling can be applied to assess the effects of accumulated depositions on ecosystems (e.g. Wright et al., 2005), and such studies require historical emission trends.

This thesis relies to a large extent on the research carried out under the LRTAP Convention and its scientifically based and policy driven program, EMEP (Co-operative programme for monitoring and evaluation of the long-range transmission of air pollutants in Europe).

Large reductions in European emissions of air pollution have been attained in the last 25 years, and this achievement can be related to the work under the LRTAP Convention and its innovative approach to design emission control at minimum cost and maximum environmental gain. The EMEP programme relies on three main elements: (1) collection of emission data, (2) measurements of air and precipitation concentrations, and (3) modelling of atmospheric transport and deposition of air pollution. The combination of these elements provides a good basis for the evaluation and qualification of European emission trends.

2 Inventory requirements

Emission inventories reported under international agreements like the Convention on LRTAP, are requested to be transparent, consistent, comparable, complete and accurate (UNECE, 2003). In this chapter we address important factors to fulfil such requirements for sulphur and nitrogen oxides inventories which are the main focus in this thesis.

2.1 Emission sources of sulphur and nitrogen oxides

Sources of emissions can be classified as *natural* or *anthropogenic*. While anthropogenic or man-made sources are a result of human activity, natural sources include biological and geological sources, wildfires and windblown dust. Sources of sulphur and nitrogen oxides are covered in the following, as these compounds are the main focus in this thesis. Volcanic eruptions and oceanic releases are the main natural sources of sulphur. Natural emissions of nitrogen oxides ($\text{NO}_x = \text{NO} + \text{NO}_2$) are released from soils and produced by lightning. Emissions from anthropogenic sources dominate substantially over natural emissions both for sulphur and NO_x . However, considerable differences in the estimates of natural emissions suggest a high degree of uncertainty. Anthropogenic global emissions of sulphur and nitrogen oxides emissions were according to Wallace and Hobbs (2006) approximately two times larger than emissions from natural sources in year 2000. The IPCC (2007a) reports global anthropogenic NO_x emissions in the 1990s to be between two and four times larger than those from natural sources. For Europe, Simpson et al. (1999) estimate anthropogenic emissions of sulphur to be 13 times higher than emissions from natural sources, and for nitrogen oxides, anthropogenic emissions are considered to be between 4 and 41 times higher than natural sources. This thesis focuses on anthropogenic

sources both because they are much larger than the natural ones, and because they are susceptible to legislative control.

Anthropogenic sources of sulphur and NO_x are usually referred to as *stationary* or *mobile*. Both categories are strongly related to *combustion* of fossil fuels, but stationary sources also include *non-combustion* (process) emissions. Anthropogenic sulphur is mainly released in the form of SO₂, and is related to the sulphur content in the fuel and retention in the combustion ash. Sulphur appears in coals as pyritic sulphur (FeS₂), organic sulphur, sulphur salts and elemental sulphur. The amount of sulphur present in coals and oils are variable, but are normally not higher than 1% and 3% sulphur by weight respectively (EMEP/CORINAIR, 2007). Nitrogen oxides are mainly emitted as NO (90-95%). There are two main mechanisms for NO_x formation in combustion processes and they are referred to as *fuel NO_x* and *thermal NO_x*. Fuel NO_x is formed when nitrogen bound in the fuel is released during combustion. This is the most important mechanism for NO_x formation in coal combustion (80-90%) and can contribute as much as 50% to the total NO_x emissions when oil is combusted. Thermal NO_x is formed by oxidation of nitrogen in air at combustion temperatures above 1600 °C, and is the main NO_x formation process in vehicles. A third mechanism, *prompt NO_x* formation, occurs only in the first, fuel rich stage of combustion and is attributed to the reaction of atmospheric nitrogen with radicals derived from the fuel.

To be considered **complete** an emission inventory should include all relevant sources. The emission sources are often aggregated in *source categories* or *sectors*, requested for reporting of emission data under national and international obligations. The contribution of each sector to the total emissions can vary considerably between pollutants, countries, over time and by inventory (e.g. Olivier et al., 1998; van Aardenne et al., 2001; Cofala et al., 2007). The global (Cofala et al., 2007) and European (**Papers I and II**, Vestreng et al., 2007 and 2008) emissions and sector distributions for sulphur and NO_x are compared in Table 1. Emissions from aviation, international shipping and open biomass burning, although relevant, are excluded from both these inventories, so they are still comparable. According to Cofala et al. (2007), the excluded sources contribute to the global NO_x emissions with about 3% for aviation, 12% for international shipping and 29% for biomass burning. The latter two sources contribute 8% and 5% respectively in the case of SO₂.

Table 1 Global and European emissions and sector distributions of anthropogenic sulphur and nitrogen oxides emissions in year 2000. (Unit: Tg SO₂ and Tg NO₂, numbers in parenthesis in %)

| | SO ₂ | | NO ₂ | |
|--------------------|-----------------|----------|-----------------|----------|
| | Global | Europe | Global | Europe |
| Power plants | 50 (53) | 12 (64) | 18 (21) | 4 (21) |
| Industry | 32 (33) | 4 (22) | 14 (17) | 3 (15) |
| Residential | 7 (8) | 2 (9) | 6 (7) | 1 (6) |
| Road transport | 2 (2) | 0.5 (2) | 34 (41) | 7 (42) |
| Off-road transport | 2 (2) | 0.4 (2) | 11 (13) | 3 (15) |
| Other | 2 (2) | 0.2 (1) | 0.4 (1) | 0.4 (1) |
| Total | 96 (100) | 18 (100) | 83 (100) | 18 (100) |

The largest emitting sectors are power plants for SO₂ and road transport for NO_x. The percentage contributions of each sector to the total emissions (shown in brackets in Table 1), are mostly comparable on the global and the European scale. This is the case for all sectors except for the two largest SO₂ emission sectors, namely, power plants and industry. This difference can be attributed to regional variability in the global inventory. While the power plant sector fully dominates over other emitting sectors in the OECD countries and the eastern European regions, the contribution from the industrial sector is comparable to the power plant sector in other areas of the world (Cofala et al., 2007, and supplementary material available at http://www.iiasa.ac.at/rains/Glob_emiss). Even larger regional differences in the sector distribution occur for NO_x, but these are compensated on the global scale to give an averaged sector distribution comparable to the European one.

2.2 Emission estimation approaches

Methodologies to calculate anthropogenic emissions of air pollution are divided in two main approaches: the bottom-up and the top-down approach (Granier et al., 2004). Emission inventories derived by any of these approaches need to be **transparent** (i.e. well documented) and **consistent** (i.e. estimates must reflect real inter annual emission differences) for all inventory years, pollutants and source categories. The bottom-up approach means that the calculations are made for individual sources, and that the total emissions are the sum of these individual sources. Bottom-up methods are generally described by the equation:

$$E_i = A_i \times (EF)_i \times (1-ER/100) \quad (1)$$

where E_i are emissions (e.g. Gg nitrogen oxides year⁻¹), A_i is the activity rate of a source (or group of sources i , for example, consumption of gasoline in passenger cars), $(EF)_i$ is the unabated emission factor (amount of emissions per unit activity, e.g. gram NO_x emitted per kilogram fuel), and ER is the overall emission reduction efficiency in percent. Additional parameters like the sulphur content of the fuels and retention are needed to estimate sulphur emissions.

Information sources of activity data and emission factors for emission estimation are shown but not limited to those listed in Table 2. The table contains an explicit ranging of the data sources that is dependent on the scope of the emission inventory work (e.g. pollutants included, time span and geographical coverage). We have separated and ranged the different information sources in terms of their reliability to develop a present day European emission inventory.

Table 2 Information sources on available activity data and emissions factors for bottom-up estimation of emissions

| | Activity data (A) | Emission factors (EF) |
|-----------------|---|--|
| National | Statistics offices Ministries | Direct measurements National research |
| Central | EMEP database (webdab.emep.int) International Environmental Agency Eurostat OECD UN Statistics Division UN Food and Agriculture Organization Manufacturers associations IIASA (iiasa.ac.at/web-apps/apd/gains) Mitchell, 1981 Darmstadter, 1971 Etemad et al., 1991 Surrogate data e.g. population | EMEP/CORINAIR Guidebook (2007) Peer reviewed publications IIASA (iiasa.ac.at/web-apps/apd/gains) Reports IPCC Guidelines (IPCC, 2006) US EPA AP-42, 1996 Data from other countries |

The national data sources listed in Table 2 are anticipated to be more reliable than the central data sources because activity data is available at the most disaggregated level. In addition, the data collector can be in direct contact with the provider of the data, and emission factors are representative for the local conditions in the country in question. The central data sources are often more readily available. Sources of activity data required to develop historical inventories are listed at the bottom of the table. The activity data inferred from population or gross domestic products (GDP) (surrogate data) are considered to be the last option, and ranged with the lowest reliability in Table 2. With respect to emission factors, direct measurements of emissions can be carried out using continuous

emission monitoring (CEM) techniques. This methodology produces the most accurate estimates, but is expensive. Such measurements are currently in use mainly at large stationary sources, which are under strict guidelines for emissions control, such as power plants. Alternatively, estimated emission factors assumed to be the same for different countries and or processes can be applied. Caution should be used when generalizing the emission factors to different regions because that may influence the accuracy of the emission estimates to the extent that they cannot confidently be applied to determine the state of the environment.

Information on abatement level per emission source and country, the emission reduction parameter (ER), is more difficult to access than other parameters in equation 1, because it is often strongly country specific and not reported to central databases. The IIASA RAINS/GAINS database (<http://www.iiasa.ac.at/web-apps/apd/gains>) and the US EPA (1996) includes information about the emission reduction efficiency (ER) of different technologies and also their level of penetration, but in other world regions, this information is not readily available and national emission calculations may rely on crude assumptions about the technology mix. Apparently, the **accuracy** (emissions are neither systematically overestimated nor underestimated) of an emission inventory generally increases with the costs involved in creating it, both because accurate measurements of emissions and emission factors are expensive, and because it is resource demanding to collect and evaluate emission factors and activity data appropriate for a particular application of the bottom-up approach.

The EMEP emission inventories, that constitutes the basis for this thesis, are to a large extent compiled as national bottom-up inventories. The national emission estimates are reported every year to the LRTAP Convention through the EMEP programme. The national emission estimates are mainly generated from the most reliable information sources given in Table 2. In some countries, detailed emission models like COPERT (<http://lat.eng.auth.gr/copert>) for the transport sector, are used to calculate emissions for some of the sectors. The advantage of applying such models is that they allow a consistent methodology to calculate emissions across different countries. EMEP data are however only available from 1980 onwards. For previous periods, the available information is sparser both for activity data and emission factors. It is our experience from the work undertaken in **Paper II** (Vestreng et al., 2008), that European activity data in distinct

consumption sectors were not readily available from international statistics prior to 1950. For non-OECD countries, fuel consumption statistics were not always available even for the period after 1950, and had to be derived from production figures. Emission factors representative for 1985 were available in the peer reviewed literature, and the first edition of the EMEP/CORINAIR Guidebook (McInnes, 1996) was a valuable source for the temporal development of emission factors in the road transport sector. However, there is a significant lack of information on emission factors for earlier periods. This implies that the accuracy of historical inventories covering periods prior to 1980 is rather low compared to the EMEP emission data for which country specific emission factors and detailed activity data are generally applied to calculate emissions.

An alternative way to derive emission estimates is through the top-down or inverse modelling approach. The top down approach is based on observations and chemical transport modelling (CTM) and these are used to constrain the budgets of chemical species and their emissions. In most applications a set of a priori emission estimates are required as first input to the transport model. These methods have been applied in atmospheric research mostly to constrain emissions of long-lived, well mixed gases, like carbon dioxide (CO₂), methane (CH₄), chlorofluorocarbons (CFC), and nitrous oxides (N₂O). For relatively short lived pollutants with high spatial variability, like SO_x and NO_x, the use of these inverse modelling approaches is more problematic. One reason for this is inherent limitations in the modelling tools to determine the spatial variability of these pollutants in air. Another reason is that the ground based observational network is insufficient to constrain the emissions of short lived air pollutants. Due to the recent increase in satellite observations of tropospheric trace gases, the top-down approach is likely to become more widely applied also for the traditional air pollutants. In a recent paper, Konovalov et al. (2008), use data from satellite instruments to investigate European NO_x emission trends by inverse modelling. Their results are in general agreement with the EMEP emission trends reported in **Paper II** (Vestreng et al., 2007), with some remarkable exceptions in Eastern Europe and Italy. These are very interesting results, as they do not only confirm the general decline in NO_x emissions in Europe over the last decade, but also highlight regions in Europe where more research is needed in order to raise the confidence in the emission data. It is however important to consider that there are limitations both in the model and in the observations when such top-down methodologies are applied to derive emission trends. While the discrepancies in results found for Eastern Europe could be attributed to the

higher uncertainty in the emission data in this region, we suspect that the lack of reduction in emissions seen for Italy in Konovalov et al. (2008) could also be due to limitations in the vertical extent of the chemical transport model applied for the calculations.

The EMEP emission trends are validated using a top-down approach on an annual basis and the results are documented in EMEP reports (e.g. Fagerli et al., 2003). Further, Jonson et al. (2006) has validated the EMEP NO_x emission trends which constitute the basis for **Paper II** (Vestreng et al., 2008). The EMEP sulphur trends in **Paper I** (Vestreng et al., 2007) are validated by the application of the Oslo CTM2 model (Sundet, 1997) and observations from the EMEP monitoring network in **Paper III** (Berglen et al., 2007). **Paper IV** (Fagerli et al., 2007) validates the work on historical emission trends for sulphur, ammonia and elemental carbon inventories based on results from the Unified EMEP model (Simpson et al., 2003; <http://www.emep.int/OpenSource>) and comparison with ice core records. These results are further discussed in chapter 3. It is far from straightforward to determine what differences between model results and observations can be explained by uncertainties in the emission estimates. However, an agreement between top-down and bottom-up approaches is a reassuring way to build up the confidence in the underlying emission estimates

2.3 Inventory requirements for model applications

Model studies undertaken to assess the impact of air pollution on ecosystems, climate and human health, requires emission data to be (1) spatially, (2) vertically and (3) temporarily distributed in a resolution which meets the objectives of the model study. Such assessments may further require a (4) speciation of pollutants like non-methane volatile organic compounds (NMVOC). Each of these requirements is addressed separately below.

1. Spatial distribution: Emissions are usually classified as line (e.g. road traffic), area (e.g. residential) and point (e.g. power plants) sources, but are often not available in spatially distributed form. In order to distribute these emissions geographically, we can apply a spatially distributed dataset which we anticipate is representative for the actual location of emissions. Such surrogate datasets are available in Europe from national institutions, as well as from central databases and satellite records. For instance, point source information is available from reporting under the Aarhus Convention PRTR Protocol, the Large Combustion Plan Directive (LCPD), and the European Pollutant Release and Transfer Register (E-PRTR). Land use data from CORINE (Coordination of information on the

environment) and satellite based land cover data derived from raw images (e.g. www.temis.nl) are useful sources of this surrogate information over Europe. For other parts of the world, global emissions are often distributed according to the EDGAR datasets (<http://www.mnp.nl/edgar> to be available at <http://edgar.jrc.it>), which rely on rural and urban population density distributions in areas outside Europe and the United States (Butler et al., 2008). A project called ECCAD (Emission of atmospheric Compounds: Compilation of Ancillary Data) (Michel et al., 2006; <http://ether.ipsl.jussieu.fr>) is now established to create a database containing global geographical data for emission estimation. The outcome of this project could in our view be very useful to further improve global emission estimates.

The basic principle for distributing emissions is presented in equation 2 using a surrogate spatial dataset (S) defined in terms of geographical coordinates (i, j):

$$E_{ij} = E_T \times \frac{S_{ij}}{\sum_{ij} S} \quad (2)$$

Where E_{ij} are the emissions attributed to a specific grid point in a particular country and E_T is the total national emission for a sector to be distributed across the country by application of the surrogate spatial dataset S. S_{ij} are the data values in each of the grid cells in the surrogate dataset that is used as a weight factor for the total, E_T , emissions. In order to create a final emission map, all the spatially distributed data per country have to be merged in a common grid.

2. Vertical distribution: In Europe, information on physical stack heights of large point sources (LPS) is reported by the Parties to the LRTAP Convention, and is also available from the CORINAIR programme under the EEA and from the GENEMIS project (Generation of European Emission Data for Episodes) (Friedrich and Reis, 2004). The GENEMIS also provides information on stack diameters and exhaust gas temperature, speed, volume and surface area for the pollutant releases. Such information is important to support plume-rise calculations to be applied to derive vertical emission distributions that can be generalised for model calculations.

3. Temporal distribution: Socio-economic data are used to describe the variation of activities and emissions on monthly, daily and hourly resolution. Examples of such

indicators for temporal distribution of emissions are fuel consumption and load curves for the power plant sector, and traffic counts in the case of road transport. For industrial processes, the production figures and working hours are important indicators, and user behaviour is recorded to estimate the temporal variation in the residential sector. Temporal profiles of emissions for the whole of Europe have been derived within the GENEMIS project (Friedrich and Reis, 2004).

4. Speciation: Different components may need more detailed information on the chemical composition of the emissions than usually provided by national emission inventories. This is the case for instance for NMVOCs, NO_x, particulate matter (PM) and mercury (Hg). In these cases, information relies on the availability of separate studies. NMVOC emissions are often only available as a sum of several hundred different species. These compounds react at different rates and by different chemical mechanisms, producing different amount of ozone. Ozone modelling thus requires the NMVOC emission to be speciated. Species profiles are available e.g. from Passant et al. (2002), Schultz et al. (2007) and Theloke and Friederich (2007). For particulate matter, information on the share of elemental to organic carbon is available for instance in Kupiainen and Klimont (2007). There is also a need to speciate the NO_x emissions, and the need for emission inventories to also include information about the share of primary NO₂ emissions is addressed in **Paper II** (Vestreng et al., 2008).

The part of the EMEP domain covered by reported spatially distributed emissions is now 32%. To complete the spatial distribution, we have developed methods in line with those outlined above and detailed in Tarrasón et al. (2004). The EMEP inventory covers emission data from 1980 and onwards. For periods prior to 1980, the spatial and vertical distributions of emissions are modified as documented in **Paper V** (Marmer et al., 2007). Further information about the vertical and temporal distribution as well as the NMVOC and NO_x speciation applied within the EMEP program is published at the internet (<http://www.emep.int/OpenSource>).

2.4 Data quality

Emission inventories are often used by atmospheric chemistry researchers, economists, national agencies and policy makers to trace compliance with agreed emission reduction obligations. These emission inventory users need to know the quality of the emission data in order to assess the robustness of their conclusions based on the emissions. Emission

inventories should therefore always be accompanied by qualitative or better quantitative estimates of the uncertainties.

Van Aardenne (2002) distinguishes between *internal* and *external* assessment of accuracy. The former type of assessment can only be applied when all information available to construct the inventory is available. Information at the internal level is not available in national submissions to EMEP. Methods to assess the quality of national emissions of air pollutants are given in the EMEP/CORINAIR Guidebook (Pulles and van Aardenne, 2004). These are applied at national level, and are available only for a limited number of countries from Informative Inventory Reports (IIR) submitted to EMEP. Thus in order to assess the quality of reported national emissions we have to rely on external assessments. These include according to van Aardenne (2002): (1) comparisons with other inventories, (2) comparisons with measurements (3) forward modelling and (4) inverse modelling.

A main achievement of this thesis is that it contains both internal and external evaluations of the quality of European emission data for sulphur and nitrogen oxides. **Paper I** includes an analysis of internal evaluation of sulphur emissions at national and sector level. **Paper II** includes an external validation of the nitrogen oxide inventory through comparison with other estimates. In **Paper III**, an external validation through comparison with other estimates is reported and in addition, top-down validation with forward modelling and ground level observations is carried out for sulphur components. In **Paper IV**, the historical emission inventories are externally validated, this time using information from ice cores.

The results from this work indicate that the uncertainty in the EMEP inventory from 1990 and onwards is below 25% both for SO_x and NO_x (**Papers I and II**, Vestreng et al., 2007 and 2008). The uncertainties for other air pollutants (Vestreng et al., 2006) as well as for individual emission sectors (Schöpp et al., 2005) are however considered to be higher. Results from the validation of the EMEP inventory by comparison with other emission estimates indicate that differences can be attributed to assumptions about the level of implementation and penetration of control measures in different countries. Abatement technologies such as Flue Gas Desulphurization (FGD) can reduce SO₂ emissions from large combustion plants by 90% (EMEP/CORINAIR, 2007). Further, three way catalytic

converters in passenger cars may reduce NO_x emission factors in average by 67% (**Paper II**, Vestreng et al., 2008).

There is still considerable work to be carried out in order to determine the accuracy of emission inventories. However, first steps have been taken to improve the quality of the emission data officially reported under the LRTAP Convention. Since 2002, there has been an increased focus on the quality of emissions reported under the Convention. The reporting guidelines (UNECE, 2003a) defines emission data quality in terms of transparency, consistency, completeness, comparability, and accuracy and the following actions have been initiated to estimate the quality of the reported emission data:

1. Formalized procedures for annual technical review of EMEP emissions were proposed in 2002 (UNECE, 2003b) and completed in 2007 (UNECE, 2007)
2. New emission reporting guidelines were developed (UNECE, 2003a)
3. Improved guidance on emission estimation (EMEP/CORINAIR, 2007)
4. Increased bilateral communication with national experts

Together these efforts have enhanced the quality and quantity of European emission data and the present thesis has benefited from this.

The number of Informal Inventory Reports (IIR) has increased by a factor 3 in four years. These reports contain information related to completeness of the inventory, methodologies used to calculate emissions and re-calculations, thus the transparency of the EMEP inventory has improved in a short time. **Comparability** (i.e. discrepancies between emission estimates are not caused by methodological differences to calculate them) has been determined by comparison of calculated implied emission factors (IEF) across countries. Values are flagged as “outliers” if there is a difference in the values of more than 10% compared with the previous and following years. In the last review of the data (Vestreng et al., 2007), below 0.5% of the calculated emission factors were flagged to be outliers. This implies that the comparability of reported data is high. Inconsistencies introduced either due to deficiency in applying homogenous methodologies over the whole emission time series or due to direct errors are also detected in the review. This has been done automatically by examination of dips and jumps in log 10-transformed emission time series by linear regression. Whether or not the identified outliers in the trend data are justified by the actual emission evolution needs to be evaluated from case to case. Manual inspection of the trends is necessary because the automatic method to detect

inconsistencies impose linearity to the trends which in most cases, and for good reasons, are non-linear. The analysis of consistency of emission trends is carried out by examining trends in the sector data, and between pollutant and country differences. Additional information to explain potential outliers in the trends is sought in IIRs and through bilateral discussions with national experts. After this process, about 10% of the time series from 1990 onwards are found to be inconsistent for SO_x and NO_x. Thus the consistency in the time series is still relatively low.

As a consequence of the last years' review of emission data, less officially reported data have been accepted for inclusion in the EMEP emission trends. For the period between 1990 and 2005, the EMEP inventory contains about 60% officially reported sector emissions of SO_x and NO_x. The coverage of reported data in the 1980s is about a factor 2-3 lower (Vestreng et al., 2007). The coverage of the time series also varies spatially across Europe, in that less officially reported data are available from Eastern Europe, Caucasus and Central Asia countries (EECCA). Officially reported data particularly in the road transport and agricultural sectors were flagged as potentially underestimated for these countries (Vestreng et al., 2007).

Up until now, formalized reviews to determine the accuracy of reported emission data has not taken place within EMEP, but procedures have now been adopted (UNECE, 2007). Formalized review procedures to evaluate the quality of spatially distributed emissions reported 5-annually from 1990 onwards have not yet been established. Simple tests of internal consistency, sector completeness and cross pollutant ratios in selected sectors has been carried out for gridded data. It is however necessary to develop further the methodologies for review of gridded data in the EMEP inventory.

3 Results

3.1 European emission trends

The main contribution from this thesis is that it provides validated long-term and historical estimates of European emissions of SO_x and NO_x. In addition, this work relates for the first time the European sulphur and nitrogen emission reductions in the last 25 years to the

implementation of specific abatement measures, as well as to the existing protocols and European legislation to control air pollution.

Global emission trends of sulphur both from pre-industrial times (Dignon and Hameed, 1989; Lefohn et al., 1999; Smith et al., 2004; Stern, 2005; van Aardenne et al., 2001) and between 1990 and 2030 (Cofala et al., 2007) have been published earlier. For Europe, historical emission estimates has been published by Mylona (1996). In addition, emission data from 1990 onwards are available from the RAINS/GAINS (<http://www.iiasa.ac.at>) and EDGAR (<http://www.mnp.nl/edgar>) databases. **Paper I** (Vestreng et al., 2007) analyses European sulphur emission trends. The main novelty of this paper is that it is the first time that long-term EMEP emission trends of sulphur from 1980 and up to the present have been compiled, reviewed, analysed and published in the peer reviewed literature. The results show that the policies to abate sulphur emissions have been instrumental in reducing emissions in Western Europe already since the 1980s. In 2004, many Western European countries have already reduced their sulphur emissions by more than 60-80% compared to 1990 levels, and the penetration of efficient measures like flue gas desulphurization and fuel switch is already high. The paper also points out important differences in the trends between Eastern European and Western European countries. The recession in the economy in Eastern Europe after 1990, is the main cause for the large emission reductions in these countries. The emission reductions in Eastern Europe in this period are a factor 1.5 higher than the emission reduction in Western Europe where reductions are mostly related to the implementation of abatement measures.

Paper II (Vestreng et al, 2008) presents the development of nitrogen oxides (NO_x) emissions since pre-industrial time, but with particular focus on the evolution of emissions from road transport and the effect of control measures taken in this sector. Global historical NO_x emission trends are available from Dignon and Hameed (1989) from 1860 to 1980. Their emission estimates are derived by regression analyses from total fuel consumption. The Dignon and Hameed (1989) emission estimates are also included in the historical inventory for Europe compiled by Schöpp et al. (2003). Global emission trends based on bottom-up calculations have been published by van Aardenne et al. (2001) for the period 1890-1990. NO_x emission trends are further available from a study by Schultz et al. (2007) between 1960 and 2000, and from Cofala et al. (2007). Emission data from 1990 onwards are available from the RAINS/GAINS (<http://www.iiasa.ac.at>) and EDGAR (<http://www.mnp.nl/edgar>) databases. The advantage of our study over existing published

historical inventories is that it applies emission factors in Europe which varies over time and by countries from 1950 onwards. This paper shows that due to the steep increase in liquid fuel consumption, emissions from road transport increased by a factor 14 between 1950 and 1980. Road transport was the main NO_x emission source in Europe already in 1970, and this sector contributes today to about 40% of the NO_x emissions. Emission increases in Western Europe in the 1980s are attributed to implementation of inefficient regulations to reduce NO_x emissions. These regulations focused instead on improving fuel efficiency and controlling carbon monoxides and hydrocarbon emissions, without a holistic view. Since 1990, however, technological abatement following new European regulations (Euro standards) has contributed significantly to the decrease of NO_x emissions in Europe, as shown in Paper II.

The specific role of technical abatement measures in the trends of SO_x and NO_x is further illustrated when analysing the historical trends of these pollutants against the trends of carbon dioxide (CO₂) emissions.

Figure 1 compares and contrasts the historical development of European sulphur (as SO₂), nitrogen dioxides (as NO₂) and carbon dioxides (CO₂). The analysis below is based on the findings from Paper I and Paper II.

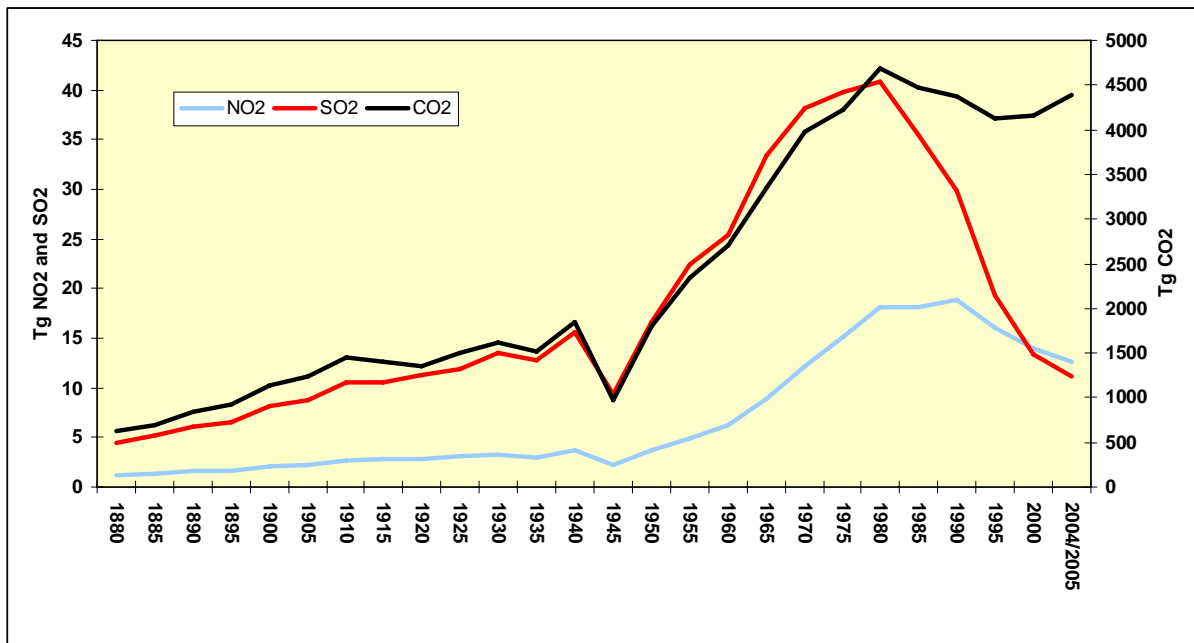


Figure 1 Trends in European emissions of SO₂, NO₂ (Mylona, 1996; Vestreng et al., 2007; 2008) (left axis), CO₂ 1880-2005 (Marland, Boden and Andres, Oak Ridge National Laboratory (<http://cdiac.ornl.gov/>) (right axis). Former USSR is excluded.

For hundred years (1880-1980) the trends in SO₂, NO_x and CO₂ are considered to be rather similar. There was a steady increase in emissions after the industrial revolution, only

broken by a sharp decrease during the Second World War, and with a much steeper trend from the 1950s and up to 1980. In this period, the emissions were mainly unabated, and thus the trends of CO₂ and SO₂ were mainly determined by the solid fuel consumption. The NO_x emission trend however relates more to the liquid fuel consumption and that is why it differs from the trends for the other two pollutants.

After 1980, the CO₂ emission trend is relatively stable compared to SO₂ and NO_x. The SO₂ emissions are reduced already in the 1980s while the emissions of NO_x do not start to decline until the 1990s. It is interesting to note that NO_x emissions have been substantially less reduced than sulphur emissions. The reasons why NO_x is less efficiently reduced as compared to sulphur can be attributed to the source distribution of emissions, to the measures available for abatement in the largest sectors and to the actual reduction efficiency. More than 60% of sulphur emissions are emitted from one single sector, namely from power plants. In order to meet obligations set forth in the UN Protocols to abate sulphur (UNECE, 2004) large sulphur reductions have been achieved, mainly from the implementation of cleansing technologies, such as flue gas desulphurization (FGD). Sulphur reductions have also been achieved by fuel switch e.g. from lignite to gas and by setting targets on the sulphur content in fuels. The largest NO_x emissions sector, road transport, contributes around 40% to the total emissions. Road transport fuels contain only negligible amounts of nitrogen thus target setting on nitrogen content is not an abatement option in this sector. About 90% of the emissions from road transport are due to reaction between atmospheric oxygen and nitrogen in the combustion air (thermal NO_x emissions). These emissions can be efficiently reduced with aftertreatment devices such as catalyst converters. The ECE-R15 regulations enforced during the 1980s resulted in improved combustion and higher NO_x emissions in road transport. This is the main reason why European reductions in this sector did not take place before requirements for catalytic converters were introduced in the 1990s by the European Commission Directive 70/220/EC. In addition to that, the efficiency in NO_x reductions from road transport is hampered both by the slow vehicle turn-over and by the increase in diesel passenger cars. This explains why the implementation of NO_x abatement measures generally requires more time to be visible in emission totals than in the case of sulphur. The above factors contribute to explain why the onset of NO_x reductions are delayed ten years compared to sulphur and also why the emission reductions from 1990 are lower.

The much lower decrease in CO₂ emissions after 1980 can be explained by the lack of possibilities for end-of-pipe measures like flue gas cleansing. Reduction of CO₂ can be obtained by decrease in consumption of fossil fuels and transition from coal and oil to less carbon-intensive energy sources, e.g. renewable energy and nuclear power. Such measures are responsible for reductions between 1980 and 2005 as depicted in Figure 1. In Western Europe the contribution of nuclear energy to total energy provision increased from 5 to 15% between 1980 and 1994. In the first part of the 1990s, reductions in CO₂ emission occurred also due to restructuring of German industries and the switch from coal to natural gas for electricity generation. In Eastern Europe, carbon dioxide emissions from fossil fuel use fell after 1990 as a result of economic recession following the break up of the Soviet Union (EEA, 1998).

Improvements in energy efficiency (e.g. better isolated houses), carbon capture and storage (e.g. in geological formations like in the Sleipner field in the North Sea) and enhancing natural sequestration (e.g. by planting more trees and reduce deforestation) are additional CO₂ abatement options. However, from 1995 onwards, the emissions of CO₂ have *increased* by 6% in Europe. According to EEA (2007a) this is mainly due to the increase in emissions from road transportation which offsets reductions in other sectors. Emission reductions of CO₂ are much more dependent on structural changes and non-technical measures (e.g. taxation) than reductions of sulphur and NO_x. For these air pollutants, large reductions have already been obtained due to technological measures. As mentioned above, technological abatement options also exist for CO₂, but the willingness to explore and implement such measures has apparently so far been more limited. The public and political awareness of climate change is rather new in comparison to air pollution, and one should not forget that the Kyoto Protocol under the UN framework Convention on Climate Change (UNFCCC) entered into force in 2005, twenty years later than the first Sulphur Protocol under the LRTAP Convention.

3.2 Top-down validation of emission trends

Another main achievement of this thesis is that it includes both internal and external evaluations of the quality of European emission data for sulphur and nitrogen oxides. The top-down validation of the emission trends contained this thesis is summarised below.

Paper III (Berglen et al., 2007) investigates trends in sulphate concentrations in Europe with the OsloCTM2 model (Sundet, 1997) and through comparison with measurements from the EMEP network. The EMEP emission trends analysed in Paper I are validated to a certain extent in this study. The paper concludes that the EMEP emission trends are reasonable, and further that the spatial distribution of emissions is better reflected in the emissions results in Paper I than in two different global inventories that are also analysed. The paper indicates that the lack of trends in emissions from shipping in the inventories hamper the comparison of model results and observations in coastal areas.

Paper IV (Fagerli et al., 2007) validates emission inventories of sulphate, ammonium and elemental carbon by model calculations with the Unified EMEP model (Simpson et al., 2003; <http://www.emep.int/OpenSource>) and comparison with observations from ice core records. This study applies the emission inventories of sulphur and nitrogen oxides presented in Paper I and Paper II but the validation corresponds mostly to the historical emission trends referred to in both papers. The research undertaken to elaborate a complete set of historic emission inventories for air pollutant gases for application in model studies is further documented in Paper V. For the study undertaken in Paper IV, two inventories for elemental carbon for the period 1920 to 1985 were developed: one based on constant emission factors taken from Bond et al. (2004) and the other based on variable emission factors in the road transport sector broadly in line with Novakov et al (2003). The conclusions from the top-down validation carried out in Paper IV are that the sulphate trends are in very good agreement with the ice core records, and that further research is needed in order to derive a reliable historical inventory of elemental carbon.

3.3 Impact of emission trends

Paper V (Marmer et al., 2007) contributes to our understanding of how the trends in sulphur emissions developed by Mylona (1996) and further in Paper I, influence the regionally and seasonally resolved evolution of the direct shortwave radiative forcing due to sulphate aerosol from 1900 to the present day. In this paper it is shown how the large reductions of SO₂ emission in Europe since 1980 have contributed to a reduction in the atmospheric sulphate load and direct sulphate forcing over Europe. It further shows that the change in sulphur emission maxima from north-west to south-east is reflected in the regional distribution of the sulphate forcing. Local responses to a radiative effect are yet uncertain, but the decline in sulphur emissions is likely to contribute to a warming over

Europe. The effect of the increase in international shipping emissions, contrasting the land based emissions, is specifically addressed in Paper V. The conclusion is that the direct radiative forcing due to emissions from ocean going ships has continuously increased from 3% in the 1980s to over 10% in 2000. This conclusion relies on year 2000 ships emissions from Whall et al. (2002). These emissions are downscaled back to 1980 by the growth rate from Endresen et al. (2003). Between 1880 and 1980, the emissions are anticipated to be directly proportional to the registered tons associated with steam and motor ships (Mitchell et al., 1981). More recently, Endresen et al. (2007) and Eyring et al. (2005) have published historical global inventories for emission from international shipping. Discrepancies both in the trends and the levels have been identified between the inventories for international shipping, and further work is needed in order to understand better the underlying causes.

For other impact assessment of European emission trends, the reader is addressed to reports in the framework of EMEP. Estimates of the impact of European air pollution on acidification, eutrophication and ground level ozone relying on the emission estimates which constitute the basis for this thesis, are published in these reports. The temporal decline in European emission levels for the last 25 years has been accompanied by reductions in the deposition of air pollutants. Large improvements in the risk damage of acidification to ecosystems have been achieved all over Europe. Model studies by Fagerli et al. (2006) show that the area of unprotected ecosystems to acidification in Europe has decreased from 41% in 1990 to 10% in 2004. Exceedances of nutrient nitrogen load have also decreased, but many countries still have more than 80% of their area at risk for eutrophication. The area unprotected to eutrophication is much larger than the area unprotected to acidification, and is less reduced (from 66% to 47%) between 1990 and 2004. According to EEA (2007b), the reduction in precursor gases (NO_x , NMVOC, CO, CH_4) have not reduced the impact of ozone on human health and ecosystems in Europe.

4 Future perspectives

The work undertaken in this thesis has contributed to quantify emissions trends of sulphur and nitrogen oxides over Europe since 1880 and to determine the quality of such estimates. It has also documented how the decrease in European SO_x and NO_x emissions in the latest 25 years is unequivocally related to the application of abatement technologies, driven by international agreements and European legislation to control these air pollutants.

In order to assess the state of the environment in the best possible way and to be able to form the most efficient policy to reduce the effects of air pollution, high quality emission inventories are required. The emission work under the LRTAP Convention has experienced noticeable progress over the last years. Aided by the development in communication systems and resources spent on bilateral communication, organization of reporting routines, guidelines, and guidance, the volume of emission data officially reported under the Convention has increased by a factor 30. Further, the focus on data quality through the annual reviews of emission data, have increased the confidence in the documented emission levels and trends. The interest to participate in the work under the Convention is also growing, and is reflected in an annual increase in the number of Parties to the Convention (fifty-one) by about one Party a year since 2000. Close collaboration with the European Commission and its European Environmental Agency (EEA), as well as the United Nations Framework Convention on Climate Change (UNFCCC) and the Intergovernmental Panel on Climatic Change (IPCC) has been established in order to harmonize the emission reporting and assure an effective evaluation of the reported data.

There is however still room for improvements. We have to continue to ask ourselves if there are sources unaccounted for, whether estimation methodologies adequately reflect the emission levels, where the largest uncertainties are, and where surprises are likely to appear. While well designed policies aid to abate emissions, further research is needed to limit uncertainties, uncover new challenges and propose solutions. This applies especially to some of the pollutants not analysed in this thesis. Limitations in emission estimation of particulate matter, heavy metals and persistent organic pollutants are reflected in the very high uncertainties associated with these pollutants (several hundred percent) (e.g. Vestreng et al., 2005). Also, ammonia emissions from the agricultural sector are not always adequately estimated. Uncertainties are due both to the lack of adequate emission factors and, particularly for non-combustion sources, methodological limitations (e.g. Breivik et al., 2006). The work to improve the emission inventory guidance (e.g. through updates of the EMEP/CORINAIR Guidebook) must therefore be looked at as a continuous effort. The technical review of emission data also needs to be developed further in order to continue to improve emission data quality. Scientifically sound methods to efficiently assure the consistency in time series and to determine the quality of spatially distributed datasets, should be explored and implemented. Extended use of reported activity data to compare

trends in implied emission factors within and across countries is one possibility to improve consistency and comparability. Methodologies to review the emission data should be designed also to facilitate quantitative feedback to the Parties and the data users about the data quality. Further, the review results should be linked to individual emission figures, and retrievable along with the emission data. In this way, the data users will be given a better opportunity to assess the uncertainty in their own results based on the emission data. Improvement in the emission data quality could also be gained through closer links between the remote sensing and emission data communities, as indicated by the top-down validation of European NO_x emission trends addressed in Paper II (Vestreng et. al., 2008).

Several different European emission inventories have been developed over the years. Most of these emission inventories are aimed at serving specific scientific and policy needs. Efforts are being made within the European ACCENT network (<http://www.accent-network.org>), to facilitate overview, access and comparison of all these inventories through development of a data portal (<http://www.aero.jussieu.fr/projet/ACCENT>). The success of this initiative to improve the access of high quality emission data for model applications is however limited due to the incompleteness and incomparability of such objective-oriented inventories. Further, information which can explain differences between inventories and favour one inventory in front of the other is often lacking. We believe that the way forward is to design projects dedicated to develop long-term emission inventories in a format suitable to serve both policy- and research-oriented applications. In our work to develop a European historical emission inventory for NO_x, we furthermore saw the need for a publicly available database with quality assured activity data from pre-industrial times, and with relevant sector detail. This database should be dynamic to include better information whenever available, and could be extended to the global scale. With such an activity database in place, resources could be liberated to be spent on other important parameters for emission calculations like the temporal evolution of emission factors.

The accuracy in the spatial distribution of emission sources needs to be further improved in order to increase our ability to confidently determine how the population and ecosystems are exposed to air pollution. Due to the increased availability of satellite observations, we foresee near future improvements in the spatial allocation of emissions data. The accuracy in the spatial distribution of emissions could also benefit from inclusion of high resolution emission data into regional and global datasets.

Where will the big surprises be found with respect to development in air pollution in the not too far future? We believe that there are plenty of opportunities for surprises in the intersections between air pollution and climate change. The reason is twofold. Firstly, according to the IPCC (2007b), human activities have already caused substantial climate change. Climatic changes may influence the already poorly understood and quantified natural emissions of air pollutants and the processes affecting them. Secondly, the air pollution and climate change research is not yet fully integrated, and many research groups are working rather separately on these two closely linked subjects. Thus, future strategies to abate global warming, might turn out to be less beneficial for air pollution (e.g. Crutzen et al., 2003; Brasseur and Roeckner, 2005; Morton, 2007). IPCC notes however that “...near-term health co-benefits from reduced air pollution as a result of actions to reduce greenhouse gas emissions can be substantial and may offset a substantial fraction of the mitigation costs” (IPCC, 2007c). This is reassuring, and our point is not to prevent action to be taken to avoid additional global warming because of potential negative effects on air pollution. Our point is that it is important to make an integrated evaluation of mitigation options in order to avoid unexpected negative side effects e.g. when alternative fuels and technologies are introduced.

The history of air pollution has taught us that it might take centuries before required legislation and technological options are in place to reduce emissions. On the other hand, when the scientific basis for international collaboration is first established, and the public, thus politicians, are willing to take the steps necessary to abate pollution, emission levels can be substantially reduced within a rather short time frame. The current goal to limit the rise in global temperature to no more than 2 °C since pre-industrial time requires a 85% reduction in global CO₂ emissions in fifty years (2000 to 2050) (IPCC, 2007d). Well aware of the different challenges related to global CO₂ abatement and European sulphur emission reductions, the 73% reduction in European SO₂ emissions in 25 years, might still make us believe that it is possible to also considerably reduce greenhouse gases if we want to.

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- UNFAO, Food and Agriculture Organization of the United Nations. <http://www.fao.org>
- UNSD, <http://unstats.un.org/unsd/energy/edbase.htm>

Appendix I: Summary of papers

Paper I:

Vestreng, V., Myhre, G., Fagerli, H., Reis, S., and Tarrasón, L.: Twenty-five years of continuous sulphur dioxide emission reduction in Europe, *Atmos. Chem. Phys.*, 7, 3663-3681, 2007.

During the last twenty-five years European emission data have been compiled and reported under the Cooperative Programme for Monitoring and Evaluation of the Long-range Transmission of Air Pollutants in Europe (EMEP) as part of the work under the UNECE Convention on Long-range Transboundary Air Pollution (LRTAP). This paper presents emission trends of SO₂ reported to EMEP and validated within the programme for the period 1980-2004. These European anthropogenic sulphur emissions have been steadily decreasing over the last twenty-five years, amounting from about 55 Tg SO₂ in 1980 to 15 Tg SO₂ in 2004. The uncertainty in sulphur emission estimates for individual countries and years are documented to range between 3% and 25%. The relative contribution of European emissions to global anthropogenic sulphur emissions has been halved during this period. Based on annual emission reports from European countries, three emission reduction regimes have been identified. The period 1980-1989 is characterized by low annual emission reductions (below 5% reduction per year and 20% for the whole period) and is dominated by emission reductions in Western Europe. The period 1990-1999 is characterised by high annual emission reductions (up to 11% reduction per year and 54% for the whole period), most pronounced in Central and Eastern Europe. The annual emission reductions in the period 2000-2004 are medium to low (below 6% reduction per year and 17% for the whole period) and reflect the unified Europe, with equally large reductions in both East and West. The sulphur emission reduction has been largest in the sector Combustion in energy and transformation industries, but substantial decreases are also seen in the Non-industrial combustion plants together with the sectors Industrial combustion and Industrial production processes. The majority of European countries have reduced their emissions by more than 60% between 1990 and 2004, and one quarter have already achieved sulphur emission reductions higher than 80%. At European level, the total sulphur target for 2010 set in the Gothenburg Protocol (16 Tg) has apparently already been

met by 2004. However, still half of the Parties to the Gothenburg Protocol have to reduce further their sulphur emissions in order to attain their individual country total emission targets for 2010. It is also noteworthy that, contrasting the Gothenburg Protocol requirements, a growing number of countries have recently been reporting increasing sulphur emissions, while others report only minor further decreases. The emission trends presented here are supported by different studies of air concentrations and depositions carried out within and outside the framework of the LRTAP Convention.

Paper II:

Vestreng, V., Ntziachristos, L., Semb, A., Reis, S., Isaksen, I. S. A., and Tarrasón, L.: Evolution of NO_x emissions in Europe with focus on road transport control measures. *Atmos. Chem. Phys. Discuss.*, 8, 10697-10747, 2008.

European emission trends of nitrogen oxides since 1880 and up to present are presented here and are linked to the evolution of road transport emissions. Road transport has been the dominating source of NO_x emissions since 1970, and contributes with 40% to the total emissions in 2005. Five trend regimes have been identified between 1880 and 2005. The first regime (1880-1950) is determined by a slow increase in fuel consumption all over Europe. The second regime (1950-1980) is characterized by a continued steep upward trend in liquid fuel use and by the introduction of the first regulations on road traffic emissions. Reduction in fuel consumption determines the emission trends in the third regime (1980 -1990) that is also characterized by important differences between Eastern and Western Europe. Emissions from road traffic continue to grow in Western Europe in this period, and it is argued here that the reason for this continued NO_x emission increase is related to early inefficient regulations for NO_x in the transport sector. The fourth regime (1990-2000) involves a turning point for road traffic emissions, with a general decrease of emissions in Europe during that decade. It is in this period that we can identify the first emission reductions due to technological abatement in Western Europe. In the fifth regime (2000-2005), the economic recovery in Eastern Europe imposes increased emission from road traffic in this area. Western European emissions are on the other hand decoupled from the fuel consumption, and continue to decrease. The implementation of strict measures to control NO_x emissions is demonstrated here to be a main reason for the continued Western European emission reductions. The results indicate that even though the effectiveness of

European standards is hampered by a slow vehicle turnover, loopholes in the type-approval testing, and an increase in diesel consumption, the effect of such technical abatement measures is traceable in the evolution of European road traffic emissions over the last 15 years.

Paper III:

Berglen, T. F., Myhre, G., Isaksen, I.S.A., Vestreng, V., and Smith S. J.: Sulphate trends in Europe: Are we able to model the recent observed decrease? *Tellus*, 59B (4), 773-786, 2007.

This model study applies three sets of emission inventories for the years 1985, 1995 and 2000 to assess if the Oslo CTM2 is capable of reproducing the steep decrease in sulphur surface trends in different European regions as seen in the emissions and observed by the EMEP measurement network. Surface concentrations of sulphate (SO_4^{2-}) are reasonably well reproduced with most station within 50% deviation, while sulphur dioxide (SO_2) is increasingly overestimated over the fifteen years period. Limitation in wintertime oxidation of SO_2 to sulphate may partly explain the differences, but further studies are needed in order to understand the results for SO_2 better. Having established that the model results for sulphate are comparable to the observations, regional trends in sulphate are studied. The modelled European decrease 1985-2000 is lower but comparable to the decline in observations (- 52% and - 59% respectively). The trend in deposition of sulphate is weaker than the trend in emissions, because relatively more SO_2 is oxidized to sulphate as emissions decreases. The study can also be looked at as a validation of the EMEP SO_2 inventory, as the model and measurement trends correspond best in most areas for this set of emissions, and further highlight the importance of the spatial distribution of emission in order to capture the observed trends. The net export of sulphur out of Europe has decreased by a factor 3 between 1980 and 2000. Sulphur deposition from upwind sources outside the continent (i.e. North America) is considered small (10%), and in addition with a downward trend comparable to the European, thus will most likely not change this result. There are indications that relatively more sulphate is transported out of Europe when emission levels drop below a certain threshold, because less oxidized SO_2 is seen to be deposited within Europe. The model results are demonstrated to be robust both with respect to meteorological variability and resolution, and indicate that a $2.8 \times 2.8^\circ$ resolution is

adequate to capture important elements as total mass and lifetime of relevant species, in addition to wind generated DMS emissions and SO₂ loss processes. Finally it is proposed that trends in emissions from international shipping might determine the observational trends in coastal regions, which makes it important to correctly reflect these trends in emission inventories.

Paper IV:

Fagerli, H., Legrand, M., Preunkert, S., Vestreng, V., Simpson, D., and Cerqueira, M.: Modeling historical long-term trends of sulfate, ammonium, and elemental carbon over Europe: A comparison with ice core records in the Alps. *J. Geophys. Res.*, 112, D23S13, doi:10.1029/2006JD008040, 2007.

The regional EMEP chemical transport model has been run for the 1920–2003 period and the simulations compared to the long-term seasonally resolved trends of major inorganic aerosols (sulfate and ammonium) derived from ice cores extracted at Col du Dôme (CDD, 4250 m above sea level in the French Alps). Source-receptor calculations have been performed in order to allocate the sources of air pollution arriving over the Alps. Spain, Italy, France, and Germany are found to be the main contributors at CDD in summer, accounting for 50% of sulfate and 75% of ammonium. In winter, when the drill site is above the boundary layer, more European wide and trans-Atlantic contributions are found. The relative impact of these sources remains similar over the whole Alpine massif although transport from US and emissions from Spain contribute less as we move eastward from CDD, toward other alpine ice core drill sites like Colle Gnifetti (CG) in the Swiss Alps. For sulfate, the CDD seasonally resolved ice core records and the simulated trends compares very well. For ammonium, the trend simulated by the model and the summer ice core record are in reasonable agreement, both showing greater changes in ammonium concentrations than would be suggested by historical ammonia emissions due to increase in emissions of SO₂ and NO_x towards 1980. Motivated by a such good agreement between simulations of past atmospheric concentrations and ice core records for inorganic aerosol species, we also use the model to simulate trends in elemental carbon for which less information on the historical evolution of emissions is available.

Paper V:

Märmer, E., Langmann, B., Fägerli, H., and Vestreng, V.: Direct shortwave radiative forcing of sulphate aerosol over Europe from 1900 to 2000. *J. Geophys. Res.*, 112, D23S17, doi:10.1029/2006JD008037, 2007.

On the basis of historical simulations of the atmospheric distribution of sulphate aerosols over Europe, we have estimated the evolution of the direct shortwave radiative forcing due to sulphate aerosol (RF) from 1900 to the present day. Following the trend in emissions and atmospheric sulphate burden the radiative forcing peaks in the 1980s. Since then, environmental policies regulating SO₂ emissions successfully reduced the atmospheric load. On average, the forcing of the year 2000, representing present day, equals that of the 1950s. Spatially, the sulphate and forcing maxima experienced a shift from the northwest to the southeast during the century. This is reflected in much stronger RF over the Black Sea today than hundred years ago, contrasting the development in other parts of Europe. The emissions from sulphur kept increasing since the 1980s, hence their relative contribution to the sulphate load and radiative forcing constantly increased, from 3% in the 1980s to over 10% in the year 2000. Forcing is strongest during summertime due to the higher availability of oxidants to convert SO₂ to sulphate, with a seasonal mean of -2.7 W m^{-2} in the 1980s and -1.2 W m^{-2} in summer 2000. The mean forcing efficiency is slightly reduced from $-246 \text{ W (g sulphate)}^{-1}$ in the 1900s to $-230 \text{ W (g sulphate)}^{-1}$ in the year 2000, due to changed geographical distribution of sulphur emissions towards areas with less solar radiation during summer.

V. Vestreng, G. Myhre, H. Fagerli, S. Reis, and L. Tarrasón. Twenty-five years of continuous sulphur dioxide emission reduction in Europe. *Atmospheric Chemistry and Physics* 2007, vol 7, pp 3663-3681.

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Twenty-five years of continuous sulphur dioxide emission reduction in Europe

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Abstract. During the last twenty-five years European emission data have been compiled and reported under the Cooperative Programme for Monitoring and Evaluation of the Long-range Transmission of Air Pollutants in Europe (EMEP) as part of the work under the UNECE Convention on Long-range Transboundary Air Pollution (LRTAP). This paper presents emission trends of SO₂ reported to EMEP and validated within the programme for the period 1980–2004. These European anthropogenic sulphur emissions have been steadily decreasing over the last twenty-five years, amounting from about 55 Tg SO₂ in 1980 to 15 Tg SO₂ in 2004. The uncertainty in sulphur emission estimates for individual countries and years are documented to range between 3% and 25%. The relative contribution of European emissions to global anthropogenic sulphur emissions has been halved during this period. Based on annual emission reports from European countries, three emission reduction regimes have been identified. The period 1980–1989 is characterized by low annual emission reductions (below 5% reduction per year and 20% for the whole period) and is dominated by emission reductions in Western Europe. The period 1990–1999 is characterised by high annual emission reductions (up to 11% reduction per year and 54% for the whole period), most pronounced in Central and Eastern Europe. The annual emission reductions in the period 2000–2004 are medium to low (below 6% reduction per year and 17% for the whole period) and reflect the unified Europe, with equally large reductions in both East and West. The sulphur emission reduction has been largest in the sector *Combustion in energy and transformation industries*, but substantial decreases are also seen in the *Non-industrial combustion plants* together with the sectors *Industrial combustion* and *Industrial production*

processes. The majority of European countries have reduced their emissions by more than 60% between 1990 and 2004, and one quarter have already achieved sulphur emission reductions higher than 80%. At European level, the total sulphur target for 2010 set in the Gothenburg Protocol (16 Tg) has apparently already been met by 2004. However, still half of the Parties to the Gothenburg Protocol have to reduce further their sulphur emissions in order to attain their individual country total emission targets for 2010. It is also noteworthy that, contrasting the Gothenburg Protocol requirements, a growing number of countries have recently been reporting increasing sulphur emissions, while others report only minor further decreases. The emission trends presented here are supported by different studies of air concentrations and depositions carried out within and outside the framework of the LRTAP Convention.

1 Introduction

Much attention has been given to the abatement of sulphur dioxide (SO₂) emissions since the 1970s, when the transboundary character of air pollutants was first robustly established and documented (e.g. OECD, 1977; Eliassen and Saltbones, 1983; Menz and Seip, 2004; Grennfelt and Hov, 2005). In order to control these emissions, international co-operation was deemed indispensable and to that purpose, the United Nations Economic Commission for Europe (UNECE) Convention on Long-range Transboundary Air Pollution (LRTAP) was established in 1979. Today, the LRTAP Convention has fifty-one Parties, forty-seven of which are European. At present, there are three international Protocols from the LRTAP Convention in force to reduce sulphur dioxide emissions. While the first Protocol, the 1985 Sulphur

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Protocol, adopted a flat rate approach (reduction of national annual sulphur emissions by at least 30% between 1980 and 1993), the two succeeding Protocols, the 1994 Sulphur Protocol and the 1999 Multi-effect Protocol (Gothenburg Protocol), are effects based (UNECE, 2004). This means that they aim at efficiently reducing sulphur emissions where environmental effects are most severe. In addition to the UN Protocols, several European Union (EU) Directives are regulating sulphur emissions, the most recent one being the 2001 National Emission Ceilings (NEC) Directive, 2001/81/EC (EC, 2001), presently under revision. The NEC Directive establishes emission ceilings to be attained by 2010 for sulphur dioxide, nitrogen oxide, volatile organic compounds and ammonia for the 25 EU Member States. Targets for the Member States that joined the EU in 2003 are specified in the Treaty of Accession (EU, 2003). In general, the sulphur emission targets for 2010 in the NEC Directive are more ambitious than those in the Gothenburg Protocol.

The main anthropogenic source of sulphur dioxide emissions is the sulphur content of fossil fuels released by *combustion*. In addition, some sulphur arises from petroleum *refining*, the smelting of sulphidic ores in the *production* of heavy metals, in the production of sulphuric acid, paper and sulphur. Natural fluxes of sulphur originate from volcanoes, and biological and photochemical production in the oceans of volatile sulphur gases, notably dimethyl sulphide (DMS). Comparably small amounts of sulphur are also emitted from forest fires, soils and vegetation, sulphur springs and sea salt (Simpson et al., 1999).

Sulphur emissions influence the level of acidification of soils and freshwater ecosystems (e.g. Stoddard et al., 1999; Schöpp et al., 2003), climate change (e.g. Haywood and Boucher, 2000; Ramanathan et al., 2001) and have impacts on human health (e.g. WHO, 2003, 2005, 2006). The acidification situation has been serious in large parts of northern Europe in the 1970s, mainly in the Fenno-Scandia region also due to slow weathering of soil and bedrock. Significant exceedances of critical loads were observed over large parts of central Europe, southern parts of Scandinavia and North-Western Europe (Lövblad et al., 2004). Emission and successive deposition of sulphur have caused material, soil and forest damage (e.g. Nellesmann and Goul Thomsen, 2001; Akselsson et al., 2004) and surface water acidification. Decreased pH and accompanying increase of aluminium compounds is fatal to fresh water fish, and in many lakes e.g. in the southern part of Scandinavia the whole fish population was completely exterminated by 1986 (e.g. Henriksen et al., 1989; Rohde et al., 1995; Yakovlev, 2001; Gunn and Sandøy, 2003; Skjelkvåle et al., 2003). A thorough review and assessment of air pollution trends and their effects was carried out with the occasion of the Convention's 25 years anniversary in 2004 (Sliggers and Kakebeeke, 2004), complemented by studies from the EMEP programme (Lövblad et al., 2004) and by the Working Group of Effects under the LRTAP Convention (WGE, 2004).

While the attention traditionally was directed towards the "acid rain" environmental problem of sulphur compounds described above, the emphasis today tends to be more on the climate and human health impacts of the particulate phase of sulphur. The most severe effects in terms of overall health burden of air pollution are associated with the long-term exposure to particulate matter. A significant reduction in life expectancy of the average population by a year or more has been estimated if present levels are to continue (WHO, 2006). In this context, the latest update of the World Health Organization (WHO) Air Quality Guidelines (WHO, 2005) reflects the need to provide a larger degree of protection against SO₂ emissions than preceding documents (WHO, 2000). Hence the limit values of 20 µg m⁻³ for 24 h average exposure and 500 µg m⁻³ for a 10-min average are much more stringent than in the 2000 revision of the Guidelines where the limit was 125 µg m⁻³ as a 24 h average (WHO, 2005). However, if SO₂ emissions should be reduced to levels which are certain to be associated with no effects, the levels would have yet to be much lower than in the current guidelines (WHO, 2005, and references therein).

SO₂ is an aerosol precursor and can be converted to sulphate aerosols. Both sulphur dioxide and sulphate have life times of less than a week, hence the influence of sulphur releases is mainly of regional character. Sulphate aerosols are shown to have a significant direct aerosol effect (Haywood and Boucher, 2000; Schulz et al., 2006) and are an important contributor to indirect aerosol effects (Haywood and Boucher, 2000; Ramanathan et al., 2001; Lohmann and Feichter, 2005). The direct and indirect aerosol effects due to sulphate lead to a negative radiative forcing and thus a cooling effect on climate. Myhre et al. (2004) showed that emission changes of SO₂ between 1985 and 1996 impact the geographical distribution of the radiative forcing of the direct aerosol effect substantially. Global anthropogenic sulphur emissions have been shown to increase rather steadily up to about 1980, but with a more uncertain trend after that (Boucher and Pham, 2002; Smith et al., 2004; Stern, 2006). The global trend is uncertain over the last decades since it consists of large reduction over North America and Europe and a large increase over Asia. Several authors have studied the possible impact on sulphur deposition due to changes in regional climate (e.g. Mayerhofer et al., 2002; Langner et al., 2005; Sanderson et al., 2006). Changes in weather pattern, temperature and precipitation has been found to both increase and to decrease acidification, imposing changes of about ±5% in sulphur deposition patterns depending on location.

In order to trace the progress in controlling transboundary air pollution and its related effects, the founding Protocol under the LRTAP Convention in 1979 agreed on the exchange of information by countries on emission data and on transboundary fluxes. The compilation of both emission data and information on transboundary fluxes has been carried out under the Cooperative Programme for Monitoring

and Evaluation of the Long-range Transmission of Air Pollutants in Europe, also named the EMEP programme. Every year since the start of the programme, EMEP has published the officially submitted information on emissions and transboundary fluxes, in recent years as well online in the EMEP emission database (<http://www.emep.int> and <http://webdab.emep.int>).

This paper presents the twenty five year trends for sulphur emissions in the EMEP area as estimated within the EMEP programme and documents the sulphur trend by country and sector in time and space. Special focus is given to the post 1990 development and the present 2004 emissions are compared with the ceilings for 2010 in the LRTAP 1999 Multi-effect Protocol (Gothenburg Protocol). Emission targets in the NEC Directive are not included, as the emission reporting requirements, hence the national total emissions reported, differs on several points between the LRTAP Convention and the NEC Directive. In addition, final NEC emission data is only complete for the EU-15 total emissions for four years (2001–2004) (http://reports.eea.europa.eu/technical_report_2006_8/en/technical_report_2006_8.pdf); hence analysis of long-term trends, which is the focus in this paper, cannot be undertaken. Emissions included here are only anthropogenic national emissions. Natural sources of sulphur are not considered here and neither are sulphur emissions from international shipping and international aviation. Although emissions from international air and sea transport may prove to contribute significantly to European air pollution assessments, these are beyond the scope of the present paper. This is the first time that the 25 years of sulphur dioxides emissions reported under EMEP are presented and analysed in peer reviewed literature. In this paper, we first discuss quality aspects of EMEP emissions data and how we work towards a complete validation of the EMEP inventory through annual review processes and by top-down assessments. The emission improvement program under EMEP has increased the transparency and confidence in official submission and is a main reason to support for the first time the publication of the EMEP trends. At the end of the paper, we discuss also the uncertainty ranges in the EMEP inventory and how the EMEP trend compares with other independent estimates and the European emission contribution to global anthropogenic sulphur inventories.

2 Emission sources and methodology

The main source of emission data used under the LRTAP Convention is national official emission reports (<http://webdab.emep.int/>, 6th version). Every year, emission data per sector from Parties to the LRTAP Convention is compiled at national level and are reported through the EMEP programme. The emission data are reported in the Nomenclature For Reporting (NFR) source categories. There are 102

NFR categories in the reporting templates (<http://www.emep.int/emis2007/reportinginstructions.html>), including both detailed categories (e.g. Residential plants, Passenger cars and Iron and Steel, in addition to Public Electricity and Heat Production) as well as the associated aggregated levels (e.g. Residential, Road Transport and Manufacturing Industries and Construction) to facilitate reporting under the Convention also for Parties with less resources available for emission estimation and reporting. Reporting according to NFR mostly applies for the 1990 and onwards emission data. The 1980s are still dominated by emission data reported in the eleven SNAP (Selected Nomenclature for Air Pollutants) source sectors as defined in the EMEP/CORINAIR Guidebook (<http://reports.eea.europa.eu/EMEP-CORINAIR4/en>), i.e. less detailed information is available for this time period. The national inventories are based on national statistics and country specific, technology dependant emission factors. National experts are requested to estimate their national emissions according to the EMEP/CORINAIR Emission Inventory Guidebook. The Guidebook offers a three Tier approach for emission estimation (<http://www.ipcc-nggip.iges.or.jp/public/2006gl> for a definition of Tiers). By moving from a lower to a higher Tier, more specific emission factors, more detailed activity information, specific abatement strategies and other relevant technical information is required. The Tier approach allows all Parties to apply the Guidebook for their emission estimation irrespectively of resources and or detail of information available for emission estimation, as emissions can be estimated on different levels of complexity. If country specific methodologies are applied, these should be documented separately. This documentation is rather scarce for the 1980s, as it is only the past few years that the requirements for national emission data have become more transparent and guidelines on emission reporting, requesting also Informative Inventory Reports (IIRs), have become available (UNECE, 2003). In addition, new routines and standards for validating emission data have recently been adopted (UNECE, 2005).

Whenever there is a lack of reported data, or the officially reported data fails to pass the quality control established in the annual review (UNECE, 2005) described in more detail below, the sector emissions are either gap-filled or replaced by independent estimates and by linear interpolation and extrapolation. The main source of information for the independent estimates is emission data from the RAINS (Regional Air Pollution INformation and Simulation) model (Amann, 2005a, b). RAINS data is currently the preferred choice, since the datasets have been thoroughly reviewed with national experts through the Clean Air For Europe (CAFÉ) programme and proved to be largely consistent and comparable with officially reported data. The methodology used to derive the RAINS emission estimates is well documented (<http://www.iiasa.ac.at/rains/caf.html>). If no data has been submitted under the LRTAP Convention, and RAINS data is not available, EDGAR emission

data (<http://www.mnp.nl/edgar/>) has been used instead, as these inventories to our knowledge are the only readily available emission inventories which covers several years of SO₂ sector data information for all European countries. In some cases, when sector data for a particular year and a particular country is missing, but data for other years are available from the country, interpolation of the values is used instead. Extrapolation of country trends is seldom required, and mostly used for the latest year when a Party has failed to submit data in time.

For the scope of this study, the emission sector data is presented according to SNAP source sectors as defined in the EMEP/CORINAIR Guidebook (<http://reports.eea.europa.eu/EMEP/CORINAIR4/en>). Sulphur emission trends prior to 1980 are taken according to Mylona (1996, 1997). The source of projected emissions data for 2010 is the emissions ceilings as stated in the Gothenburg Protocol (UNECE, 2004) supplied with RAINS scenario data (Amann, 2005a, b).

3 Validation of the EMEP emission data

3.1 The review process

For the last three years, new routines to evaluate and improve the quality of emission data officially reported under the Convention on LRTAP and the National Ceilings Directive (NEC) have been established under the EMEP programme in collaboration with the European Environmental Agency (EEA) and its European Topic Centre on Air Quality and Climate Change (ETC-ACC). The review is based on the key parameters *Transparency, Consistency, Comparability, Completeness and Accuracy* as defined in the Emission Reporting Guidelines from UNECE (2003).

The estimation and validation of European emission data is first facilitated through the continuous development of the EMEP/CORINAIR Guidebook. The Emission Inventory Guidebook (<http://reports.eea.europa.eu/EMEP/CORINAIR4/en>) assists the national experts in their emission estimation work and is intended to reflect the best available knowledge on methodology and choice of emission factors for all components and sectors required for reporting. Whenever updates of the Guidebook become available that may affect the estimation of a certain pollutant, the Parties are requested to recalculate the whole emission time series in order to secure methodologically *consistent* emission time series. The use of a common methodological framework also aims to assure *comparability* between national emission inventories, and adds to the transparency of the inventories.

The review of the national emission estimates is presently organised according to recent routines established in UNECE (2005). The review of emission data consists of three stages;

1. **Stage I** checks the *timeliness* and *format* of submissions;

2. **Stage II** evaluates key sources (IPCC, 2000) and establishes the *completeness, consistency, comparability* and *transparency* of reported data, and
3. **Stage III** involves an in-depth review by individual countries which aims at establishing the actual *accuracy* of the emission estimates.

Annual Stage I and II emission data reviews are performed by a review team of experts. These reviews have been performed by EMEP since 2004 in collaboration with ETC-ACC and the UNECE secretariat, covering both data reported under the NEC Directive and under the Convention on LRTAP. Each Member State and Party is provided with a country specific review report three months after the data submission, summarizing conclusions from the review and the bilateral discussions with individual country emission experts necessary for clarification of certain aspects of the emission data. Countries are encouraged to give explanations for or correct data within the next reporting round. Meanwhile, if an explanation is not found, a replacement of country data by independent estimates is undertaken. As mentioned also in Sect. 2, replacements might simply consist of linear interpolation between two adjacent years. Whenever outliers are identified several places in the inventory, it is necessary to replace the whole time series by independent estimates, as the latter rarely will be fully consistent with the emission data provided by the country itself. General conclusions from the review are subsequently documented in an annual joint EMEP/EEA review report (e.g. Vestreng et al., 2006a). In addition, well organized reporting routines have proven to be crucial in the work of inventory improvement under EMEP. Thus, upfront the annual submission of data, reporting instructions detailing the requirements as laid down in the Emission Reporting Guidelines (UNECE, 2003) together with template files for reporting are made available online. Further to that, an online QA/QC tool, REPDAB (Vestreng, 2003), checking the *completeness* and *consistency* of reported emission data has been developed and made available to countries for quality control of data before submission.

Despite the short time since the initiation of the new emission data review routines, there has been considerable improvement in the quality of the reported emission estimates. The results from Stage I reviews indicate that from 2004 to 2006 the number of reports submitted within deadline has increased by 50%, thus improving the *timeliness* of the inventory. It is also an achievement that emission data are now reported to EMEP in the agreed file structure and according to the agreed Nomenclature For Reporting (NFR) formats. Aggregated sectors are generally consistent with the more detailed categories reported. This is thought to be mainly due to the set up of the reporting routines, the availability and use of REPDAB and the review team's increased focus on the importance of consistency for the review process. The improvements made under the Stage I review facilitate considerably the review tasks under Stage II below.

The transparency and the availability of additional information concerning the compilation of the national emission inventories has dramatically increased thanks to the active involvement of national experts in the review. The number of Informative Inventory Reports (IIR) with detailed information on the methodologies used to compile emissions and justification of changes with respect to the EMEP/CORINAIR Guidebook has increased threefold in the last three years. Also the number of bilateral consultations with national experts and the number of replies to the country specific review reports has considerably increased. This means that the *transparency* of emission data as defined in the Emission Reporting Guidelines (UNECE, 2003), has greatly improved over a relatively short time period.

Differences amongst countries due to differences in emission estimation methodologies and reporting guidelines are assessed through five *comparability* tests in the Stage II review. The tests include recalculations, inventory comparisons (NEC or United Nations Framework Convention on Climate Change (UNFCCC) reported data versus LRTAP data comparisons), implied emission factor (IEF) and cross pollutant checks, as well as the basis for transport emission calculations (fuel used versus fuel sold). Results which fall outside the empirical ranges of averages are identified as outliers by the review team of experts, thus possibly an error, which could however well be explained by national and source-specific circumstances not familiar to the review team. The review team seeks to find explanations for defined outliers in the submitted IIRs, based on knowledge within the expert review team and through country replies to its review reports. The conclusion so far is that most countries follow the methodologies in the EMEP/CORINAIR Guidebook although different Tiers are used in different countries, implying that the quality and comparability of the inventories are not fully homogenous throughout the whole EMEP area.

The *consistency* and *completeness* of reported time series of emission data per sector is crucial in trend studies of air pollution. As stated before, Parties are encouraged to submit complete inventories and recalculate the whole time series of emissions whenever new information becomes available. However, sometimes only the latest years or data back to 1990 are recalculated by applying best available methodologies and emission factors, while the remaining part of the time series may consist of data reported according to SNAP source categories. The fact that an inventory consists of reported data in different formats does not necessarily mean that the data are not consistent according to the emission data review. The review team analyses the consistency of all the reports by testing the behaviour of the time series for each particular sector in each individual country. Outliers are defined as dips and jumps in the time series depending on sector and pollutant and flagged for potential replacements necessary to be performed in order to guarantee consistency over time.

For SO₂ about 30% of the reported sector data for each year has to be replaced by independent estimates and about 10% of the required emissions are not submitted. This implies that officially reported and accepted sector emissions cover only 60% of the total sulphur trend time series from 1990 until present (Vestreng et al., 2006b). Completeness in the 1980s has not yet been quantified, but is known to be lower (e.g. Vestreng et al., 2005). The completeness of the time series varies also spatially across Europe, with a larger lack of officially reported data in Eastern Europe, Central Asia and Caucasus countries. This is indicated in Table 1 where countries are presented in four groups ranging on the level of completeness of the reported and reviewed time series of SO₂ emission data. The first group is highlighted in grey and corresponds to the nineteen countries that have a complete and consistent official report of sulphur emissions for all years since 1980. The second group of countries represents the countries where data has been reported for most of the sectors and years but with gaps that needed to be filled. These are a total of thirteen countries and are indicated with bold italics. The third group of countries have not reported any or only fragmentary official estimates and for these RAINS estimates, interpolation or extrapolation have been used instead. These are Albania, Bosnia and Herzegovina, Luxembourg, the Russian Federation, Serbia and Montenegro, The Former Yugoslav Republic (TFYR) of Macedonia and Turkey, a total of seven countries indicated in Table 1 with stars behind the country names. The last group of countries are those that have not reported any or very little official estimates and for which RAINS data estimates are not available, so that EDGAR data have been used instead. These are: Armenia, Azerbaijan, Georgia, Iceland and Kazakhstan, a total of five countries indicated in Table 1 in normal font.

It is worth noting that replacements are never used for compliance checking performed under the LRTAP Convention, but merely to assist in atmospheric transport calculations and impact assessments. Feedback from the national experts themselves and from the review team of experts indicate that the emission data improve through the review process, but so far it has been difficult to directly quantify the improvements, mainly because the Emission Reporting Guidelines (UNECE, 2003) does not give clear guidance regarding what criteria to review against, and in addition, some of the review tests have been altered or added from one year to the next. The review is done for each reported pollutant, and for some pollutants like particulate matter and pesticides, the EMEP/CORINAIR Guidebook does not provide sufficient information. Conclusions on the review of reported persistent organic pollutants can be found for example in Breivik et al. (2006).

Additionally to the sector totals, spatially distributed emissions are necessary for modelling the dispersion of sulphur pollution. The completeness of official reports of spatially distributed sector data is lower than for the sector totals. Gridded sector data is requested in five-yearly intervals from

Table 1. Sulphur trends per European country 1980–2004 (Unit: Gg SO₂). Countries highlighted in – Grey: Officially reported data. Bold italics: Reported data completed by independent estimates. Stars: RAINS data, interpolation and extrapolation. Normal: EDGAR data, interpolation and extrapolation

| | 1980 | 1985 | 1990 | 1995 | 2000 | 2004 |
|----------------------------|--------------|--------------|--------------|--------------|--------------|--------------|
| Albania* | 72 | 73 | 74 | 14 | 32 | 32 |
| Armenia | 141 | 100 | 86 | 15 | 11 | 8 |
| Austria | 344 | 179 | 74 | 47 | 32 | 29 |
| Azerbaijan | 603 | 543 | 615 | 262 | 162 | 130 |
| Belarus | 740 | 690 | 888 | 344 | 162 | 97 |
| Belgium | 828 | 400 | 361 | 262 | 171 | 154 |
| Bosnia and Herzegovina* | 482 | 483 | 484 | 360 | 420 | 427 |
| Bulgaria | 2050 | 2314 | 2007 | 1477 | 918 | 929 |
| Croatia | 150 | 164 | 178 | 70 | 60 | 85 |
| Cyprus | 28 | 35 | 46 | 41 | 51 | 45 |
| Czech Republic | 2257 | 2277 | 1876 | 1090 | 264 | 227 |
| Denmark | 450 | 333 | 176 | 133 | 27 | 23 |
| Estonia | 287 | 254 | 274 | 117 | 96 | 90 |
| Finland | 584 | 382 | 259 | 95 | 74 | 83 |
| France | 3216 | 1496 | 1333 | 968 | 613 | 484 |
| Georgia | 230 | 273 | 43 | 6 | 7 | 5 |
| Germany | 7514 | 7732 | 5289 | 1708 | 630 | 559 |
| Greece | 400 | 500 | 487 | 536 | 493 | 537 |
| Hungary | 1633 | 1404 | 1011 | 705 | 486 | 240 |
| Iceland | 18 | 18 | 9 | 9 | 9 | 9 |
| Ireland | 222 | 140 | 186 | 161 | 131 | 71 |
| Italy | 3437 | 2045 | 1795 | 1320 | 755 | 496 |
| Kazakhstan | 639 | 575 | 651 | 528 | 506 | 425 |
| Latvia | 96 | 97 | 97 | 47 | 10 | 4 |
| Lithuania | 311 | 304 | 263 | 92 | 43 | 40 |
| Luxembourg* | 26 | 26 | 26 | 7 | 4 | 4 |
| Malta | 29 | 29 | 29 | 33 | 26 | 17 |
| Netherlands | 490 | 258 | 189 | 127 | 72 | 66 |
| Norway | 136 | 91 | 53 | 34 | 27 | 25 |
| Poland | 4100 | 4300 | 3278 | 2381 | 1507 | 1286 |
| Portugal | 266 | 198 | 317 | 332 | 306 | 203 |
| Republic of Moldova | 308 | 282 | 175 | 94 | 13 | 15 |
| Romania | 1055 | 1255 | 1310 | 882 | 727 | 685 |
| Russian Federation* | 7323 | 6350 | 6113 | 3101 | 2263 | 1858 |
| Serbia and Montenegro* | 406 | 478 | 593 | 428 | 396 | 341 |
| Slovakia | 780 | 613 | 542 | 239 | 127 | 97 |
| Slovenia | 234 | 241 | 198 | 127 | 99 | 55 |
| Spain | 3024 | 2542 | 2103 | 1809 | 1479 | 1360 |
| Sweden | 491 | 266 | 117 | 79 | 52 | 47 |
| Switzerland | 116 | 76 | 42 | 28 | 19 | 17 |
| TFYR of Macedonia* | 107 | 109 | 110 | 93 | 90 | 87 |
| Turkey* | 1030 | 1345 | 1519 | 1397 | 2122 | 1792 |
| Ukraine | 3849 | 3463 | 3921 | 2342 | 1599 | 1145 |
| United Kingdom | 4838 | 3714 | 3699 | 2343 | 1173 | 833 |
| Grand Total | 55340 | 48448 | 42896 | 26282 | 18263 | 15162 |

1990 onwards, but only twelve Parties to the LRTAP Convention have reported gridded sector data of any vintage in the 50×50 km² EMEP grid by 2006 (<http://www.emep.int/grid/>).

These countries represent 24% of the emissions and 25% of the area covered by the Parties listed in Table 1. EMEP is thus required to account for the spatial distribution of

emissions for a large part of Europe by applying its own methods. The methodology for allocating SO₂ emissions is mainly dependent on the distribution of large point sources, but additional information is also necessary on vertical and temporal variation of emissions (Tarrasón et al., 2004). Review of gridded sector data is also performed under EMEP, but is at present not formalized to the same extent as for the emission totals and is out of the scope for this study.

3.2 Validation of European SO₂ emission trends using measurements and modelling

In addition to the review of emission data mentioned above, top-down assessments that use both measurements and modelling of air concentrations and depositions are of significant importance to increase the confidence in emission data. It is well known that the sulphur components can be transported over long distances, thus the validation of emission trends cannot be judged by comparing measurements in a region with local emissions. Moreover, the processes that determine the distribution between the different sulphur components (e.g. sulphur dioxide, sulphate and the amount of sulphur deposited dry and wet) may change depending on the chemical composition of the atmosphere as well as meteorological conditions. For instance, the oxidation of sulphur dioxide to sulphate depends on the availability of oxidants, and in the late 1970s to early 1980s when SO₂ emissions peaked, the amount of oxidants was a limitation for the conversion of SO₂ to sulphate, especially in winter (Fagerli et al., 2003; Roelofs et al., 1998). At present, larger proportions of SO₂ are converted to sulphate. This has led to a smaller decrease in sulphate concentrations than in SO₂ emissions. Moreover, whilst SO₂ emissions have decreased dramatically, ammonia emissions have remained at the same level (Vestreng et al., 2005). As a consequence the dry deposition of SO₂ has become more efficient over the years, as the surface acidity to a large extent governs the resistance to dry deposition (Fowler et al., 2001). Finally, year-to-year variations both in air concentrations and wet deposition are large, e.g. of the order of 20% for sulphate (van Loon et al., 2005), and thus long time series are needed in order to detect trends in observations.

In order to use measurements to validate the emission trends, it is important to know in what direction and to what extent the trends derived from measurements could be expected to deviate from the emission trends. Furthermore, model simulations may indirectly be used to validate emission trends by comparing the model output to measurements, providing that they incorporate the important processes. SO₂ and sulphate background concentrations have been monitored in Europe at several sites since around 1980, for instance through the EMEP Programme. Lövblad et al. (2004) assessed the trends in EMEP emissions and measured atmospheric concentrations and deposition of sulphur compounds in Europe from the end of the 1970s until 2000. For SO₂, they found national reductions in SO₂ emissions and

average reductions in SO₂ measurement concentrations at national sites to correspond well. Both measurements and emissions changed around 90% for countries like Germany, United Kingdom, Sweden, Austria, Finland and Denmark. For Czech Republic, Italy, Latvia, Lithuania and Switzerland differences between the reductions in the national EMEP SO₂ emission inventories and the change in average SO₂ concentrations was in the order of 5%. A somewhat larger difference was found in Belarus and Slovakia (emission reductions of 80–85%, average SO₂ concentration reductions of 60–65%), possibly due to the location of the sites. Measurement sites situated at high altitudes, near large sources in neighbouring countries or downwind of large indigenous sources do not necessarily show the same trend as the national emission. In general, however, agreement between national emission reductions and concentrations at sites in a country for the primary component SO₂ are better than for the secondary component, sulphate, that are transported over longer distances. For sulphate, the decrease was found to be less than for SO₂ (typically 50–70%), consistent with the higher conversion rate to sulphate during this period as discussed above. The same pattern was found for oxidized sulphur in precipitation, probably because sulphate particles are the main contributor to oxidized sulphur in precipitation. In conclusion, the trend in the measurement data was found to support the reported trend in emissions.

Model simulations can also be used to validate emission changes indirectly through comparison with measurements, however, only a few studies aiming at comparing long term trends in measurements with modelled trends have been performed. Berglen et al. (2006) modelled 1985, 1995 and 2000 using the EMEP and Smith et al. (2004) inventories, together with the combined GEIA (<http://geiacenter.org>)/EDGAR (<http://www.mnp.nl/edgar/>)/Aerocom (Dentener et al., 2006) dataset. They were able to reproduce the sulphate trends to a large extent, although the model showed a slightly smaller decrease (52%) than the observations (59%) for the 1985 to 2000 period using the EMEP inventory. Sulphur dioxide was increasingly overestimated over the years from 122% (modelled concentrations are in average more than twice as large as observed) in 1985 to 349% in 2000. Although the modelled and measured decreases for sulphate 1985–2000 correspond within a few percent, indicating that the trend in the emission data is reasonable, it is difficult to conclude more specifically on the quantity of the emission trend, as the trends in the primary component SO₂ (which are closely related to the emission changes) are not very well reproduced in their model simulations.

Evaluation of sulphur trends in air and precipitation using the EMEP Unified model have also been carried out using the EMEP inventory (Fagerli et al., 2003). In this study, 9 different years were calculated (1980, 1985, 1990, 1995–2000), taking into account also the annual meteorological variability. The sulphate trends were well reproduced, with a deviation between model results and EMEP measurements (on

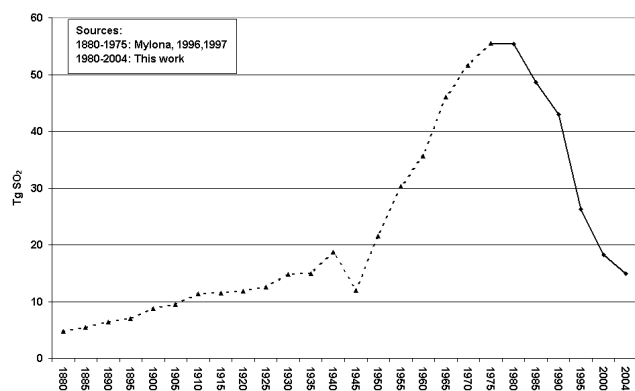


Fig. 1. Historical development of sulphur dioxide emissions in Europe (Unit: Tg SO₂).

average) between -10% and 6% for the different years. Similar to Berglen et al. (2006), the decrease in SO₂ concentrations between 1980 and 2000 was too low compared to the decrease in observations. Whilst modelled SO₂ concentrations in 1980 were in good agreement with the observations (on average overestimated by 4%), the model simulations overestimated SO₂ by 39% in 2000. In the EMEP Unified model, a parameterisation of the so-called co-deposition of NH₃ and SO₂ is implemented in the dry deposition module (Simpson et al., 2003). If this effect is not taken into account, the overestimation of SO₂ around 2000 is even larger (around 80%), whilst it has little effect around 1980 when SO₂ emissions peaked (H. Fagerli, personal communication, 2007). Thus, a part of the increasing overestimation of SO₂ as calculated by Berglen et al. (2006) can be explained by the rather simple dry deposition scheme in their model. It is not clear why the EMEP model still overestimate SO₂ for recent years. However, the major part of the reduction in the SO₂ concentrations are captured by the model simulations, hence the trend in the EMEP SO₂ emission inventory does correspond to the observed decreases in SO₂ concentrations.

4 Results

4.1 Historical changes in sulphur emissions

The EMEP inventory's twenty-five years of sulphur dioxide emission decreases are presented below in a long-term perspective. Emission data from Mylona (1996, 1997) is included prior to 1980 since these emission were comparable with the EMEP inventory for overlapping periods. The historical development of sulphur emissions since 1880 are presented in Fig. 1, based on Mylona (1996, 1997) for the period 1880 to 1975 and on the EMEP inventory which is based on official reported emission data, from 1980 and onwards. From the pre-industrial area to the outbreak of the Second World War the European SO₂ emissions were in-

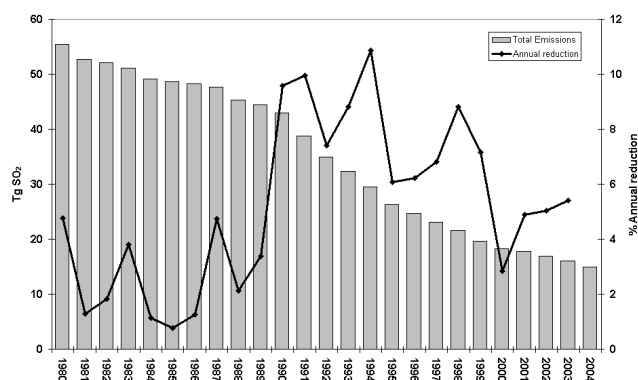


Fig. 2. Total sulphur dioxide emission trend (Unit: Tg SO₂) and relative annual European emission reduction 1980–2004 (Unit: %).

creasing slowly but steadily from 5 to 19 Tg SO₂ as a result of increase in power generation from solid fuels. The emissions decreased to World War I level during the World War II, but thereafter grew steeply to about 55 Tg SO₂ in the late 1970s due also to the availability of liquid fuel to satisfy the increase in energy demand. During the last twenty-five years, European SO₂ emissions have decreased rapidly to 15 Tg SO₂, and have in 2004 reached the same level as 70 years ago. Figure 2 (grey bars) presents a closer look at the last twenty-five years of sulphur reduction in Europe. The reduction since 1980 has been significant (73%). The reasons for these considerable reductions are a mixture of the economic situation, implementation of abatement technologies, restructuring of energy sources at national level and increased awareness of the need to reduce sulphur emission through existing international instruments as the Protocols to the LRTAP Convention discussed in some detail below. The emission trends per country as tabulated in Table 1 indicate that while the European SO₂ emission trend has been continuously decreasing during the whole period 1980–2004, the emission trends vary considerably between individual countries. In each one of the five-years periods listed in Table 1, there are countries with nearly constant emissions, countries which increase their emissions and others with emission decreases. There are also large variations in the size and location of the emission changes, and these differences are analysed here.

The key sources for SO₂ are the sectors *Combustion in energy and transformation industries*, *Non-industrial combustion plants*, *Combustion in manufacturing industries* and *Production processes*. In Fig. 3 we have plotted the trends in emissions from these sectors from 1990 to 2004. The sector trends are gradually decreasing and flattening out towards 2004. Largest reductions have been obtained from *Combustion in energy and transformation industries*, followed by *Non-industrial combustion plants* and *Combustion in manufacturing industries*. The relative contribution to total SO₂ emission from the *Combustion in energy and*

transformation industries has increased slightly on behalf of the *Non-industrial combustion plants* sector. However the key sectors remain the same during the whole period.

Figure 2 displays both absolute and relative annual European emission reductions. Largest relative reductions (black curve) took place in the beginning of the 1990s, with a maximum in 1994–1995 (11%). Largest absolute reduction (grey bars) is seen between 1990 and 1991. The annual reduction was smallest in absolute terms between 2000 and 2001, while the relative reductions were only 1% between multiple years in the 1980s. Based on the annual relative emission reductions, we have identified three emission regimes. The annual sulphur reduction is shown to be below 5% in 1980–1989 (Low reduction regime) and largest (up to 11%/year) for 1990–1999 (High reduction regime). The downward emission trend has flattened out from 2000 onwards, and annual reductions for the five year period, 2000–2004 (below 6%/year), are almost back to 1980s level (medium-low reduction regime). In the first reduction regime, 1980–1989, the total European reduction was 11 Tg or about 20%. Total reduction in the second ten year period was more than twice as large (23 Tg or about 54%). In the last 5 year period a reduction of only 3 Tg (17%) could be observed.

The spatial disaggregation of emissions for the three reduction regimes is presented in the difference maps 1980–1990, 1990–2000, 2000–2004 in Fig. 4. The picture shows that the countries responsible for the emission reduction during the low reduction regime (Fig. 4a), were mainly Western European, notably Germany, France, Italy, United Kingdom and Spain. Some of the Eastern European countries like the Russian Federation, Poland, Hungary and Czech Republic, did also reduce their emissions, but characteristic for this period is the Eastern European increase of emissions (Fig. 4a). Western European countries were responsible for about 75% of total European emission reduction in this reduction regime. During the high reduction regime (Fig. 4b), most Eastern European countries decreased their emissions considerably, notably the Russian Federation, Ukraine, Czech Republic, Poland, Bulgaria and Belarus. The Eastern European countries were responsible for about 80% of total European reductions during the high reduction regime. Only Turkey continued to substantially increase the sulphur emissions in this period. Finally, the most recent reduction regime reflects the unified Europe with about equal, and medium to small reductions in both Western and Eastern Europe as shown in Fig. 4c. Slight increases are seen also in this period in the Former Yugoslavian countries, Greece, the Republic of Moldova, Bulgaria and Finland.

The three reduction regimes defined above are closely connected to the individual countries' political and economical development. In the early 1980s, when the environmental problems connected to acidification were confidently highlighted, no supranational instruments were in place to help policy makers forming long-term strategies to abate emissions. In addition few countries had the economical and

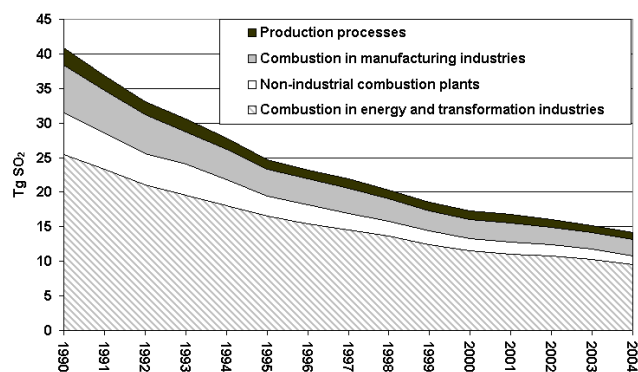


Fig. 3. Sector trends for of sulphur dioxide emission key sources 1990–2004 (Unit: Tg SO₂).

technological ability to implement the required measures, and this is reflected in the relatively low European reductions between 1980 and 1999. This situation was dramatically changed in the following ten year period where one sulphur protocol already was in place, the second Protocol was adopted, and work was ongoing in order to prepare for the Gothenburg Protocol. While the Western European countries continued to implement new technologies and fuels in order to meet Protocol targets and reduce the identified environmental problems, the economic recession in Eastern Europe, resulting in a drop in activity level, had a larger overall effect on the emission reductions. From 1995 the activity level in many Eastern European countries stabilized and started slowly to increase. However the emissions kept dropping as measures, in particular *Flue Gas Desulphurization* (FGD), but also extensive fuel switches from solid/liquid to gaseous fuels were implemented. Lately, the tendency is towards an increase in activity level in both East and West, but the total emissions have not increased yet due to the high penetration of emission control technology.

We have analyzed further our three reduction regimes by comparing trends in fuel consumption with emission trends. It should be noted that, while Parties to the LRTAP Convention are reporting activity data which is linked to the reported emissions on a five-yearly basis from 1990 onwards, fuel consumption data post 1990 consistent with the reported emissions have not been readily available for this study. For the period 1980–1990 we have studied trends in international statistics of solid fuels (UNECE, 1983, 1985b, 1990, 1992). Neither of these sources of fuel consumption data is as complete in terms of temporal resolution and coverage as are the emission data, and a comprehensive analysis by country and sector of the causes for the emission reduction pattern data is not attempted here.

In Eastern Europe, we find that the changes in solid fuel consumption and emissions follow each other closely between 1980 and 1990, while the picture is more dispersed in Western Europe. In Western Europe, emission reductions

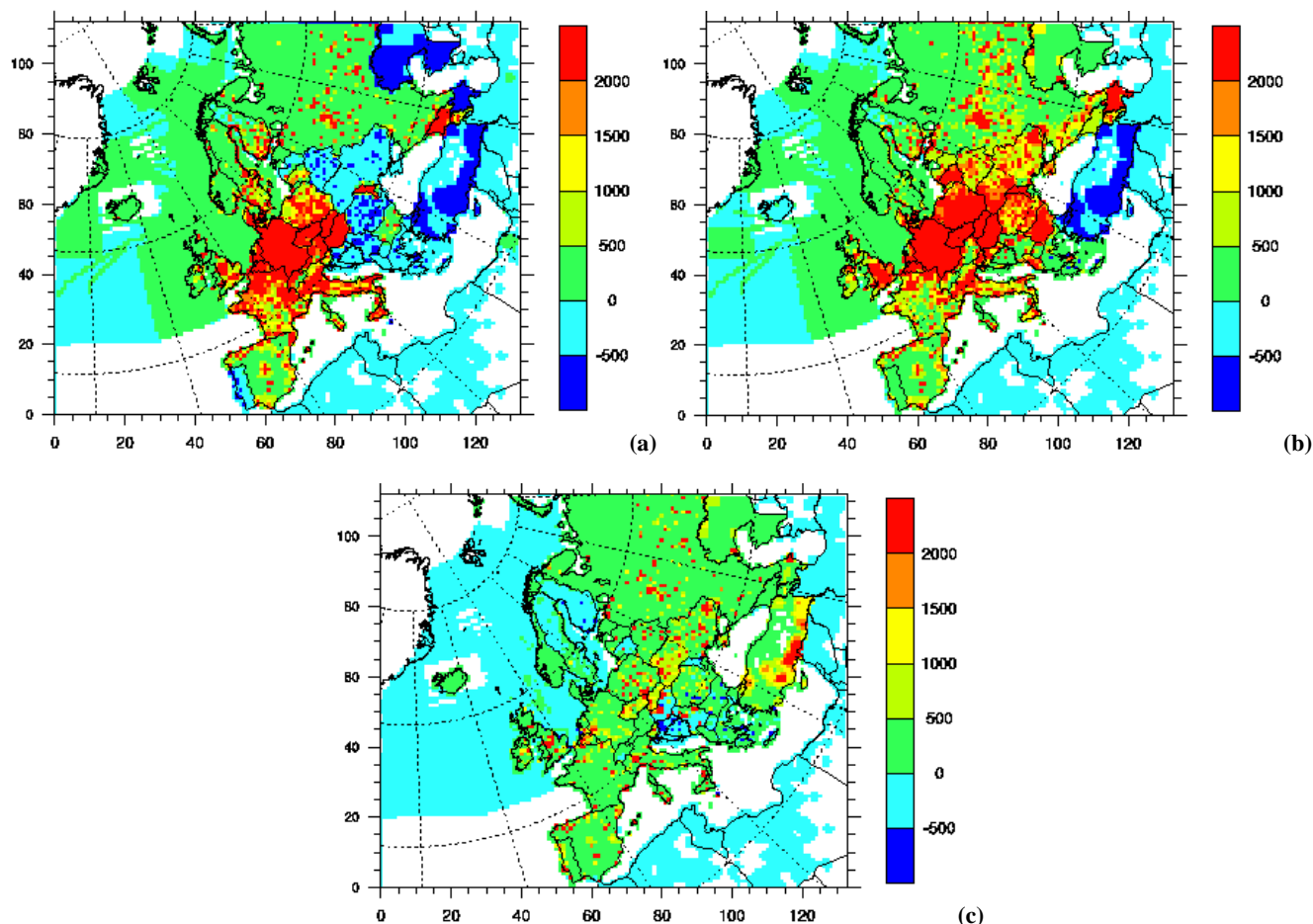


Fig. 4. Difference maps presenting the three European SO₂ emission reduction regimes. Reductions between 1980 and 1990 (a: Upper left), reductions between 1990 and 2000 (b: Upper right) and reductions between 2000 and 2004 (c: Bottom). Unit: Mg SO₂/grid cell.

have been possible also thanks to policy regulations already from the early 1980s, leading to implementation of advanced emission control technologies such as Flue Gas Desulphurization (FGD) (mainly in power generation), together with fuel switch (e.g. from coals with a high sulphur content to gas and or nuclear power) and increased sulphur removal from refined petroleum products, and not because the demand for sulphur producing activities (energy and heat production, industrial and residential combustion) has ceased. Hence, a clear decoupling of SO₂ emissions from the trend in activities has been observed already in the first reduction regime in Western Europe.

The period between 1990 and 2000 was dominated by the Eastern Europe socio-political changes and resulting in the transition from a centrally planned economy to a free-market economy. The structural changes were accompanied by a significant drop in industrial production, hence also energy consumption. The resulting decrease in energy production is directly reflected in corresponding emission reductions of sulphur in countries with the largest reductions during this period like Poland and the Czech Republic, as is also pointed out by Mill (2006). Both in Eastern and Western Europe, the

reduction in solid fuel consumption were compensated by increased consumption of gas, renewable and nuclear energy particularly in the last part of this period.

The emission abatement strategy in Former East Germany (GDR) and Former West Germany (FRG) is a good example on how policies, implementation of measures and structural changes are closely linked to the emission reduction pattern and how it has been possible to decrease German (GDR and FRG) emissions by 18% in the 1980s (1980–1989) and by 85% in the ten year period (1990–1999) following the reunification. In the FRG the reduction of SO₂ emissions was forced by the implementation of the Federal Emission Pollution Control Act in year 1974 and by several following Federal Emission control ordinances. These regulations caused a wide spread implementation of highly efficient emission control technologies, as well as a switch from solid fuels like coal and lignite to oil and gas, and increased use of low-sulphur heating oil, and resulted in a gradual drop in FRG sulphur dioxide emissions already from 1974 onwards. On the opposite side, and due to financial restrictions, the economy in the GDR was based to the extent possible on the use of domestic lignite (e.g. in 1989 more than 70% of the total

primary energy consumption was based on the combustion of lignite). In the years leading up to 1990, there was no specific regulation for the use of domestic lignite and – besides a few very small tentative facilities and, started in 1987, one power plant in Berlin – no advanced emission control technologies were in place in the GDR. Hence, the sulphur dioxides emissions in the three largest sectors in GDR, *Combustion in energy and transformation industries*, *Combustion in manufacturing industries* and, *Non-industrial combustion plants*, increased until the end of the 1980s. Since the German reunification in 1990, the reduction of SO₂ emission has been dominated by the replacement of old facilities by new ones with the best available technology and regulation for desulphurisation of flue gases in large combustion plants in the eastern part of Germany together with a fuel switch from solid to gaseous and liquid fuels. Since 2001 the national government encouraged the consumption of low-sulphur fuel in the road transportation by a tax discount.

Many countries have already taken necessary steps to substantially decrease their emissions of sulphur. Table 2 shows that by 2004, more than half of the countries have reduced their emissions by 60% or more of the 1990 values, and one quarter of the countries have reduced emissions by more than 80%. Only two countries report increases in the emission level (Turkey and Greece), which can be explained by growth rates of their economies and the related increasing demand for energy. The absolute reductions obtained between 1990 and 2004 are largest for Germany, the Russian Federation, United Kingdom and Ukraine. Reductions estimated for these countries are larger than the reductions from all the other European countries together and has a pronounced impact on the sulphur deposition pattern in Europe as shown in Fagerli et al. (2006).

It should be noted, however, that there is now an increasing number of countries reporting increased emissions from one year to another to EMEP. The development from the early 1990s when the reductions were largest, and up to present is shown in Fig. 5. The overall trend is clearly increasing, despite some fluctuation. More than 30% of the European countries reported increased emissions of sulphur from 2002 to 2003. This is more than three times as many as ten years earlier. Some of Europe's largest emitter countries have increased their sulphur emissions from 2000 onwards for the first time since the mid eighties. For most countries, notably the Eastern European countries in which the economy is now recovering, the increase is due to increased public electricity and heat production. The Scandinavian countries, however, report increases in emissions from national shipping. The stabilization of the European emission trend (Figs. 2 and 4), the large reductions already achieved by many countries (Tables 1 and 2) and the growing number of countries reporting annual emission increases (Fig. 5), is a worrying development of the sulphur emission trend in Europe, and it needs to be closely monitored and further assessed.

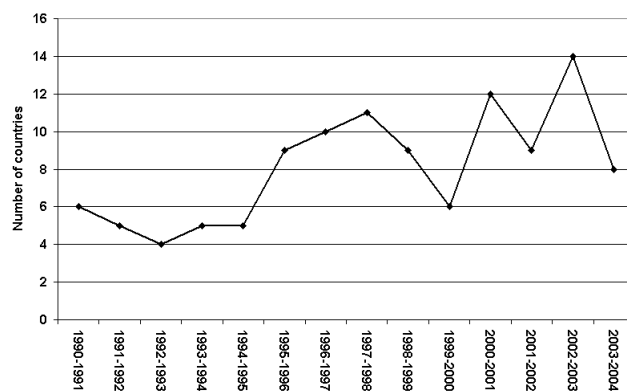


Fig. 5. Number of countries with emission increases from one year to another.

4.2 Comparison of 2004 SO₂ emission data with targets given by the Gothenburg Protocol

Table 2 shows the level of attainment in 2004 of the Gothenburg Protocol 2010 emissions ceilings. Officially reported emissions for 1990 and 2004 completed as outlined in Sect. 3.1 are listed, together with the 2010 emission ceilings, the percentage reduction from 1990 attained by 2004, and in the last column, the percentage emission reductions between the 1990 base year emissions and the 2010 ceilings as listed for information purposes in the Protocol. The table groups the European countries in three different groups depending on their status relative to the Gothenburg Protocol (UNECE, 2004) and presents the reductions obtained by Parties to the Protocol (i.e. those eighteen European countries which had ratified the Gothenburg Protocol by summer 2006), Signatories and “Other” countries. While Parties and Signatories to the Protocol together with Belarus and Cyprus have 2010 emissions ceilings listed in the Protocol, we have included in Table 2 the 2010 estimates from the RAINS model (Amann et al., 2005b) for remaining countries, including the Russian Federation which only has ceilings for its Pollutant Emissions Management Area (PEMA) listed therein.

On a European level the target for SO₂ reduction set by the Gothenburg Protocol has apparently been attained in 2004. As shown at the bottom of Table 2, the total European emissions in 2004 were about 15 Tg SO₂, while the sum of 2010 emission targets is 16 Tg SO₂. Reductions by individual countries are however still expected to be achieved, as half of the Protocol Parties have to reduce their emissions further to attain the 2010 target established by the Gothenburg Protocol (Table 2). Likewise, both the Signatories and “Other” countries groups have already attained their total 2010 targets of 3.9 and 7.3 Tg SO₂ respectively. Based on a comparison between 2004 emissions and Protocol Parties’ targets for 2010, the largest near future European reductions should take place in the Western part of Europe, notably in Spain and the United Kingdom since these two countries alone must reduce

Table 2. Level of attainment in 2004 of the Gothenburg Protocol 2010 emission ceilings. Officially reported emissions for 1990 and 2004 are listed, together with the 2010 emission ceilings, the percentage reduction from 1990 attained by 2004, and the percentage emission reductions between the 1990 base year emissions and the 2010 ceilings as listed for information purposes in the Protocol.

| | 1990 Gg SO ₂ | 2004 Gg SO ₂ | 2010 Gg SO ₂ | 2004–1990 % | 2010 % |
|---|----------------------------|----------------------------|----------------------------|----------------|--------------|
| <i>Parties to the Gothenburg Protocol</i> | | | | | |
| Bulgaria | 2007 | 929 | 856 | –53.7 | –57.0 |
| Czech Republic | 1876 | 227 | 283 | –87.9 | –85.0 |
| Denmark | 176 | 23 | 55 | –86.9 | –70.0 |
| Finland | 259 | 83 | 116 | –68.0 | –55.0 |
| Germany | 5289 | 559 | 550 | –89.4 | –90.0 |
| Latvia | 97 | 4 | 107 | –95.9 | –10.0 |
| Lithuania | 263 | 40 | 145 | –84.8 | –35.0 |
| Luxembourg | 26 | 4 | 4 | –84.6 | –73.0 |
| Netherlands | 189 | 66 | 50 | –65.1 | –75.0 |
| Norway | 53 | 25 | 22 | –52.8 | –58.0 |
| Portugal | 317 | 203 | 170 | –36.0 | –53.0 |
| Romania | 1310 | 685 | 918 | –47.7 | –30.0 |
| Slovakia | 542 | 97 | 110 | –82.1 | –80.0 |
| Slovenia | 198 | 55 | 27 | –72.2 | –86.0 |
| Spain | 2103 | 1360 | 774 | –35.3 | –65.0 |
| Sweden | 117 | 47 | 67 | –59.8 | –44.0 |
| Switzerland | 42 | 17 | 26 | –59.5 | –40.0 |
| United Kingdom | 3699 | 833 | 625 | –77.5 | –83.0 |
| <i>Total Parties</i> | <i>18 563</i> | <i>5257</i> | <i>4905</i> | <i>–71.7</i> | <i>–74.0</i> |
| <i>Signatories to the Gothenburg Protocol</i> | | | | | |
| Armenia | 86 | 8 | 73 | –90.7 | 0.0 |
| Austria | 74 | 29 | 39 | –60.8 | –57.0 |
| Belgium | 361 | 154 | 106 | –57.3 | –72.0 |
| Croatia | 178 | 85 | 70 | –52.2 | –61.0 |
| France | 1333 | 484 | 400 | –63.7 | –68.0 |
| Greece | 487 | 537 | 546 | 10.3 | 7.0 |
| Hungary | 1011 | 240 | 550 | –76.3 | –46.0 |
| Ireland | 186 | 71 | 42 | –61.8 | –76.0 |
| Italy | 1795 | 496 | 500 | –72.4 | –70.0 |
| Poland | 3278 | 1286 | 1397 | –60.8 | –56.0 |
| Republic of Moldova | 175 | 15 | 135 | –91.4 | –49.0 |
| <i>Total Signatories</i> | <i>8964</i> | <i>3405</i> | <i>3858</i> | <i>–62.0</i> | <i>–56.0</i> |
| <i>Other countries</i> | | | | | |
| Albania | 74 | 32 | 30 | –56.8 | –59.5 |
| Azerbaijan | 615 | 130 | 15 | –78.9 | –97.6 |
| Belarus | 888 | 97 | 480 | –89.1 | –25.0 |
| Bosnia and Herzegovina | 484 | 427 | 411 | –11.8 | –15.1 |
| Cyprus | 46 | 45 | 17 | –2.2 | –15.0 |
| Estonia | 274 | 90 | 44 | –67.2 | –83.9 |
| Georgia | 43 | 5 | 9 | –88.4 | –79.1 |
| Iceland | 9 | 9 | 29 | 0.0 | 222.2 |
| Kazakhstan | 651 | 425 | 237 | –34.7 | –63.6 |
| Malta | 29 | 17 | 12 | –41.4 | –58.6 |
| Russian Federation | 6113 | 1858 | 2464 | –69.6 | –59.7 |
| Serbia and Montenegro | 593 | 341 | 277 | –42.5 | –53.3 |
| TFYR of Macedonia | 110 | 87 | 82 | –20.9 | –25.5 |
| Turkey | 1519 | 1792 | 1708 | 18.0 | 12.4 |
| Ukraine | 3921 | 1145 | 1457 | –70.8 | –48.0 |
| <i>Total other countries</i> | <i>15 369</i> | <i>6500</i> | <i>7272</i> | <i>–57.7</i> | <i>–48.0</i> |
| Grand Total | 42 896 | 15 162 | 16 035 | –64.7 | –61.0 |

their emissions by 794 Gg SO₂. A closer look at those countries with remaining reduction obligation towards 2010 reveals that individual Parties to the Gothenburg Protocol are further away from attaining their emission ceilings than the Signatory countries and countries without commitments towards the Protocol. The total emission reductions required by the Parties (956 Gg) is more than five times higher than the sum of the reductions still required by the Signatories (176 Gg) as indicated in Table 2. “Other” countries will also have to reduce their emissions substantially towards 2010 in order to obtain the targets as listed in Table 2 (553 Gg), but we must take into account that neither these countries nor the Signatories have firm binding obligations under the Gothenburg Protocol.

5 Discussion

5.1 Evaluation of uncertainties

In general, the uncertainty of SO₂ emissions in the Eastern European countries is larger than for Western European countries because the level of reporting and resources available for in-country quality control is more limited. An additional factor to consider when determining the uncertainty of the emission trends presented in this paper is that the quality of the data varies also in time as some countries only recalculate their time series back to 1990 when improvements in estimation and measurement methodologies become available. In addition, the review and the work on completing the time trends has up to now mostly focussed on post 1990 emissions. Emissions before 1990 might therefore be attributed higher uncertainty than more recent data.

The increased reporting by countries on air pollutant uncertainties in their Informative Inventory Reports (IIR) in combination with uncertainty analysis of LRTAP gases published elsewhere, encouraged a first tentative quantification of the uncertainty in the EMEP SO₂ inventory. Uncertainty assessments of greenhouse gases (GHG) for the EU-15 countries were combined (based on Tier 1 estimates from 13 Member States) for the first time in the European Commission National Inventory Report (NIR) 2005 (EC, 2005). The overall uncertainty for all GHG was shown to be 4–8% measured as 95% confidence intervals. Lowest uncertainty was found for stationary fossil fuel combustion (1%). The EMEP inventory is also a compilation of emissions from different sources as pointed out in Sect. 2. The largest part of the EMEP inventory consists of emission data officially reported under the Convention on LRTAP complemented by RAINS emission estimates. We do not have as good coverage of individual countries' uncertainty analysis of air pollutants as is available for the GHG, and are not attempting to provide a complete uncertainty analysis of the EMEP inventory. We present here a compilation of relevant published uncertainty estimates of both officially submitted data (Vestreng et

al., 2006b, and references therein) and of RAINS estimates (Schöpp et al., 2005). Uncertainty estimates of air pollutants calculated by Parties rely on the IPCC Good Practice Guidance (IPCC, 2000) adopted for LRTAP gases by Pulles and van Aardenne (2001). Two different methods for uncertainty quantification are recommended therein; a Tier 1 error propagation approach and a Tier 2, stochastic simulation (Monte Carlo) analysis. A Tier 2 approach would make more sense for the quantification of uncertainty in SO₂ emissions as significant dependencies and correlations may exist particularly for fossil fuels (IPCC, 2000). However, Van Gijlswijk et al. (2004) show that for the Netherlands there were no substantial differences between Tier 1 and Tier 2 results for SO₂. Seven Parties have published their uncertainty estimates. Finland, Norway and United Kingdom have applied Monte Carlo analysis while Denmark, France and Czech Republic rely on the Tier 1 approach. The Netherlands provide both Tier 1 and Tier 2 estimates. The officially reported uncertainty estimates in total SO₂ emissions in the Western European countries is rather low and of the order of 3–7%. Uncertainty in the Czech Republic however was estimated to be about five times larger (Vestreng et al., 2006b). In order to complete the EMEP inventory, modelled emission estimates from the RAINS model (<http://www.iiasa.ac.at/rains/>) are included, particularly for some of the Eastern European countries. The uncertainty in the modelled SO₂ RAINS emission estimates are calculated based on methods specifically developed to analyse the uncertainties in RAINS estimates, considering also the uncertainties in the model parameters themselves. Generally higher uncertainties are found for RAINS estimates than for the officially reported data, and with a typical range of 10 to 15% (Schöpp et al., 2005). According to Schöpp et al. (2005), data for some Central and Eastern European countries are more uncertain than for the EU-15 countries, but the uncertainty do not exceed $\pm 23\%$ for any country. We therefore conclude that the uncertainty in the post 1990 emission estimates for individual countries included in the EMEP inventory lies between 3% and 25%, implying that the uncertainty in the EMEP inventory as such is lower. Emission data before 1990 might be subject to higher uncertainties as indicated above. Uncertainty in RAINS sector emissions is about twice as large as for the national total due to the more limited potential for error compensation (Schöpp et al., 2005).

In recent years, the lowermost uncertainty level is equally large to the annual European sulphur reduction (Fig. 2). Still, even with their inherent uncertainty, the twenty-five years trends presented here are supported by both models and measurements, and has been accompanied by reported improvements and recovery of adverse effects. Fagerli et al. (2006) shows that there has been a major reduction in the risk damage of acidification to ecosystems all over Europe from 1990–2004. While 40% of Parties to the Gothenburg Protocol had their ecosystems at risk in 1990, the unprotected area has decreased to 20% in 2004 (Fagerli et al., 2006).

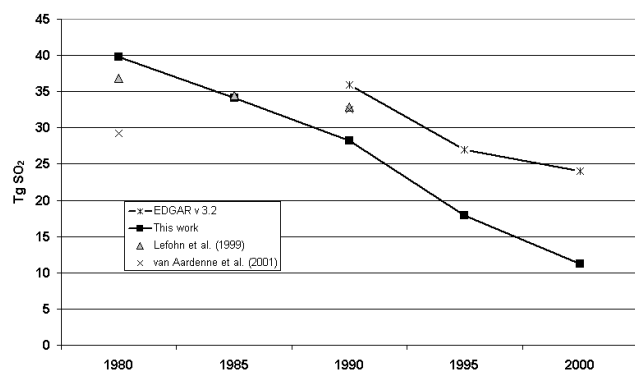


Fig. 6. Comparison of SO₂ inventories for Europe 1980–2000 (OECD Europe and Eastern Europe, excluding former USSR countries, Turkey and Cyprus) (Unit: Tg SO₂).

Skjelkvåle et al. (2005) report also widespread improvement in surface water chemistry since 1990. Last but not least, the increased effort by the Parties to the LRTAP Convention to develop good in-country QA/QC systems, followed by enhanced transparency in emission estimation methods and uncertainties documented in the IIRs, together with the ongoing emission review process, allows for an improved confidence in the officially reported emissions used for air quality and health impact modelling.

5.2 Comparison with other anthropogenic SO₂ emission estimates

The share of European emissions (EMEP inventory) to global anthropogenic emissions has been reduced from 40% in 1980 to 18% in 2000. This implies that the European contribution to global anthropogenic sulphur emissions has more than halved over the last two decades. The global estimates referred to here are those compiled and estimated by Stern et al. (2006). It should be noted that while estimates of global sulphur emission estimates compare well in level and trend up to 1950, relatively larger differences may occur particularly after 1980 (e.g. Lefohn et al., 1999; Olivier and Berdowski, 2001; Smith et al., 2001). A comparison between the most recent global total sulphur inventories by Smith et al. (2004) and Stern (2006) between 1980 and 2000 shows however that they compare surprisingly well taken into account the differences in estimation methodology applied in most areas. The global total estimates of sulphur emission differ by less than 5% between 1980 and 1992 while the differences increase to 12% (6.5 Tg SO₂) in 2000, Stern (2006) estimating lower emissions than Smith et al. (2001). Both Smith et al. (2004) and Stern (2006) include the EMEP inventory for Europe and are hence excluded from the following comparison with this inventory.

Our comparison is therefore focussed to the three inventories of Lefohn et al. (1999), van Aardenne et al. (2001) and EDGAR version 3.2 inventory (Olivier and Berdowski,

2001) as presented in Fig. 6. The EMEP inventory is the only of these inventories covering the whole time span of interest (1980–2004), so the comparison is carried out there where data from the other inventories is available. For the period 1980 to 1990 the annually resolved inventory from Lefohn et al. (1999) and the ten yearly resolved data per region from van Aardenne et al. (2001) are included in the comparison. Between 1990 and 2000, the five yearly resolved EDGAR inventory (Olivier and Berdowski, 2001) were included. The two latter inventories are related as they are both based on the same constant 1990 SO₂ emissions factors provided by J. Berdowski as personal communication (Aardenne et al., 2001) in addition to international statistics. The emission factors provided by Berdowski are country-specific and based on the sulphur contents of different fuels and metal ores and country-specific sulphur recovery by smelters, refineries and FGD in power generation (J. Olivier, personal communication, 2007). Lefohn et al. (1999) base their calculation on national and international statistics for the extraction and production of sulphur bearing fuels and metals, sulphur content in those fuels and varying degree of Flue Gas Desulphurization (FGD) control. In contrast, the EMEP inventory is based on country specific technology dependent emission factors and both national and international activity statistics. In order to make sure that we compare emissions from identical areas, we decided to exclude emissions from the territory of the Former USSR, Turkey and Cyprus, and concentrate the comparison on the areas “OECD Europe” and “Eastern Europe” as defined in EDGAR.

A year by year comparison between the EMEP inventory and Lefohn et al. (1999) inventories (not shown in Fig. 6) between 1980 and 1990 shows that these two inventories compare well in level and trend up to 1984, Lefohn et al. (1999) estimating around 5% lower emissions. Thereafter Lefohn et al. (1999) estimate an increase in emissions between 1984 and 1986 and coincide with the EMEP inventory in 1985. Lefohn et al. (1999) emissions are relatively constant between 1986 and 1989 and decrease with the same gradient as the EMEP inventory between 1989 and 1990. Both Lefohn et al. (1999) and the EMEP inventory show an overall emission decrease between 1980 and 1990, but the Lefohn et al. (1999) emission reduction is rather small compared to EMEP (Fig. 6). The difference between the inventories is three times larger in 1990 than in 1980, and might be attributed to difference in applied emission control, a factor which becomes increasingly important with time for the accuracy of emission estimates.

Van Aardenne et al. (2001) indicate an increasing trend in sulphur emissions between 1980 and 1990, opposing the trend in both Lefohn et al. (1999) and the EMEP inventory (Fig. 6). The reason for this is an increase in emissions in Eastern Europe. Since the emission factors are kept constant, this increase should be due mainly to increases in the consumption of solid fuels, as discussed in Sect. 4. The main difference between the inventories seems to be that the

1990 emission factors applied to estimate the van Aardenne et al. (2001) 1980 emissions were low compared to those applied in the EMEP and Lefohn et al. (1999) inventory.

We see from Fig. 6 that for the year 1990 all the inventories included in the comparison have relatively similar emissions estimates. The difference between the EDGAR and the EMEP inventories is 28%, while the van Aardenne et al. (2001) and Lefohn et al. (1999) total European estimates for 1990 coincide. The comparison made between the van Aardenne et al. (2001) and Lefohn et al. (1999) global inventories in the paper by van Aardenne et al. (2001) shows that also the 1990 global total estimates are the same. Possible explanations for this similarity in emission estimates at both global and regional scale are not discussed in van Aardenne et al. (2001), and it is not possible for us either to conclude if this is a mere coincidence or an indication of a better accuracy in these estimates. Best comparability was anticipated to be found between the EDGAR and the van Aardenne (2001) emission estimate for 1990 since the emission factors used in these inventories are the same. It seems however that e.g. difference in the activity data and or the more refined sector split in EDGAR give higher 1990 emissions for Europe than van Aardenne et al. (2001). Between 1990 and 2000 both the EDGAR and the EMEP emissions for Europe are strongly reduced, but the trends are flattening out towards year 2000. The EDGAR emission estimates are highest throughout the whole ten year period. The difference between the inventories increases with time, and particularly the last five years. Attention should be paid to the fact that by the year 2000 the difference in SO₂ emission estimates between the two inventories is as large as the EMEP total European emissions. The increasing difference between the inventories may be attributed to the lack of technology dependent emission factors in the EDGAR inventory, and the comparison with our work tentatively quantifies the importance of this dependence towards year 2000.

6 Conclusions

The emissions estimates presented here are compiled and validated under the EMEP programme as part of the work under the Convention on Long-range Transboundary Air Pollution (LRTAP). They conclude that European SO₂ emissions have dropped by 73% between 1980 and 2004. Reductions of sulphur emissions have been largest in the *Combustion in energy and transformation industries* sector, and reductions have been obtained both due to policy regulation followed by implementation of measures, and to economic recession. It is shown that the sulphur emission reductions were largest in the 1990s. Three distinct emission regimes have been identified. During the first period from 1980 to 1989 emission reductions were generally low (20%), and largest in Western Europe. The highest emission reductions were reported in Eastern European countries during the second period, 1990-

1999, characterised by high emission reductions (54%). The unification of Europe has led to a more equally spread reduction pattern, with low-medium reductions all over Europe, and a total reduction of 17% in the first half of the 2000s.

Our analysis of the reasons behind the emission changes in Europe shows that the policy development plays an important part in order to reduce emissions. While the Eastern European changes in fuel consumption is directly reflected in the sulphur emissions during the first reduction regime, the Western European emissions are already decoupled from the fuel consumption thanks also to policy regulations already from the early 1980s, leading to implementation of advanced emission control technologies and changes in the fuel mix notably from coal to oil and gas. During the second reduction regime, the economic recession in Eastern Europe and accompanying drop in activity level, had a factor 1.5 larger effect than policy measures taken in the western part on the overall European sulphur reduction. Recent increases in fuel consumption in the recovering economies in Eastern Europe and also in Western Europe, is mainly from fuels which do not contain appreciable amount of sulphur. In addition, penetration of control technology all over Europe is reflected in a small but continuous decrease in European sulphur emissions.

The amount of officially reported emissions to EMEP which can be confidently used in trend studies vary both in time and space and this is reflected in the uncertainty of the EMEP inventory. Eastern European emission inventories and emission estimates prior to 1990 are considered to have associated the highest uncertainties. Further, we tentatively conclude that the uncertainty in the total SO₂ emissions is between 3% and 25% for individual countries and years comprising the EMEP inventory, implying that the uncertainty in the EMEP inventory as such is lower. The EMEP emission data is subject to national QA/QC documented in Informative Inventory Reports, external annual inventory reviews, and has in addition been validated by models and measurements that support the reported trends (Lövlad et al., 2004; Fagerli et al., 2003). The sulphur emission reductions have been accompanied by a widespread improvement in surface water acidity and exceedances of critical loads¹ (WGE, 2004; Skjelkvåle et al., 2005; Fagerli et al., 2006). The reduced sulphate concentrations over Europe have led to a reduction in the cooling effect of sulphate aerosols. Local responses to a radiative effect are yet uncertain (Hansen et al., 2005). However, with such a strong reduction in the SO₂ emissions it is likely that this has contributed to a warming of Europe.

According to the EMEP estimates, the European contribution to the global anthropogenic sulphur emissions has more

¹The basic idea of the critical load is to balance the deposition rate to an ecosystem with its long-term capability to buffer the input or to remove it without harmful effects inside or outside the system (Hettelingh et al., 2001; UBA, 2004)

than halved over the last two decades. The EMEP inventory has been compared with other independent estimates provided by Lefohn et al. (1999), van Aardenne et al. (2001) and EDGAR version 3.2 inventory (Olivier and Berdowski, 2001). The downward trend over Europe is steeper in the EMEP inventory than in all the other inventories. This is probably due to the fact that the EMEP inventory uses more detailed country specific emission factors and includes changes in technology mixes. From 1990 onwards the importance of capturing the changes in technologies by applying emission factors reflecting national circumstances becomes more evident in the comparison. The EMEP inventory shows an increasingly larger emission reduction between 1990 and 2000 than the EDGAR inventory which is based on constant 1990 emission factors. It should also be noted that by the year 2000 the EDGAR estimate of total European emissions is more than twice as large as the EMEP estimate.

After the stabilization of the European SO₂ emissions since 2000, when many countries have already achieved emissions reductions of 60–80% with respect to 1990 levels, an increasing number of countries have started to report increases in national emissions. This is a worrying development that needs to be further assessed, especially as it contradicts the expectations from the target setting of existing international instruments to reduce sulphur emissions. From the perspective of the Gothenburg Protocol, further reductions should be expected, particularly in Western Europe. The Protocol target seems to be reached on a European level already by 2004, but half of the Parties have not yet fulfilled the requirements therein. In the longer term, larger reductions from Eastern Europe could be expected as several Eastern European countries have as of yet not reached an abatement level reflecting the state of the art in control technologies available. Emissions from international shipping, is not considered in this paper but clearly also influence the air quality and climate (e.g. Derwent et al., 2005; Marmer and Lange-mann, 2005). The trends in the ships emissions contrast the land based trends by an estimated increase of about 2.5% annually on cargo and 3.9% on passenger vessels in European waters (Cofala et al., 2007; Vestreng et al., 2006). Moreover, the SO₂ emissions from shipping are projected to increase by 42% from 2000 to 2020 in the current legislation baseline scenario (Cofala et al., 2007). Depending on ambition level, the emission from ships might stabilize or even decrease by as much as 70% in the case where maximum technological feasible reductions are considered. Regional differences in trends are expected, as the sulphur emissions in the Baltic Sea and the North Sea are now regulated by the MARPOL Convention Annex VI (MARPOL, 1978) which entered into force in 2005.

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Evolution of NO_x emissions in Europe with focus on road transport control measures

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Abstract

European emission trends of nitrogen oxides since 1880 and up to present are presented here and are linked to the evolution of road transport emissions. Road transport has been the dominating source of NO_x emissions since 1970, and contributes with 40% to the total emissions in 2005. Five trend regimes have been identified between 1880 and 2005. The first regime (1880–1950) is determined by a slow increase in fuel consumption all over Europe. The second regime (1950–1980) is characterized by a continued steep upward trend in liquid fuel use and by the introduction of the first regulations on road traffic emissions. Reduction in fuel consumption determines the emission trends in the third regime (1980–1990) that is also characterized by important differences between Eastern and Western Europe. Emissions from road traffic continue to grow in Western Europe in this period, and it is argued here that the reason for this continued NO_x emission increase is related to early inefficient regulations for NO_x in the transport sector. The fourth regime (1990–2000) involves a turning point for road traffic emissions, with a general decrease of emissions in Europe during that decade. It is in this period that we can identify the first emission reductions due to technological abatement in Western Europe. In the fifth regime (2000–2005), the economic recovery in Eastern Europe imposes increased emission from road traffic in this area. Western European emissions are on the other hand decoupled from the fuel consumption, and continue to decrease. The implementation of strict measures to control NO_x emissions is demonstrated here to be a main reason for the continued Western European emission reductions. The results indicate that even though the effectiveness of European standards is hampered by a slow vehicle turnover, loopholes in the type-approval testing, and an increase in diesel consumption, the effect of such technical abatement measures is traceable in the evolution of European road traffic emissions over the last 15 years.

10698

1 Introduction

The historical trend in the anthropogenic emission levels of nitrogen oxides ($\text{NO}_x = \text{NO} + \text{NO}_2$) is increasingly important for our understanding, hence our ability, to optimize abatement of air pollution and reduce the adverse effects of these pollutants on ecosystems, human health and climate, on local, regional and global scales.

The anthropogenic NO_x emissions are dominated by combustion processes in road transport with a 40% share in 2005, followed by power plants (22%), industry (16%), off-road transport (15%) and the residential sector (7%) (Vestreng et al., 2007a). Anthropogenic emissions in Europe are at least four times larger than the natural emissions from lightning, soil emissions and forest fires (Simpson et al., 1999). European anthropogenic emissions of NO_x contribute to about 30% of global NO_x emissions in 1990, when excluding ships and biomass burning (Olivier et al., 1998; Cofala et al., 2007; Vestreng et al., 2006; Schultz et al., 2007). The evolution of emissions in Europe in the last 15 years (1990–2005) contrasts with the situation in Asia, Latin America, Middle East and Africa, where less policy regulations are in place and NO_x emissions are increasing (Naja et al., 2003; Cofala et al., 2007).

Much effort has already been invested in order to abate NO_x emissions in Europe, both at national and at European-wide level. The first UNECE regulations to control emissions from motor vehicles (ECE-R15) were already being discussed in the 1950s and came into force in 1970 (UNECE, 1958; Berg, 2003). They were designed to reduce the emissions of carbon monoxide (CO) and hydrocarbons (HC) due to incomplete combustion. The early European legislation can be viewed as a response to the US initiatives, which had at that time already introduced air pollution control policies to address the degradation of air-quality in Los Angeles, California. Much later, and within the framework of the Convention of Long-range Transboundary Air Pollution (LR-TAP), two Protocols regulating NO_x entered into force; the 1988 Sofia Protocol sets a limit to national annual emissions or transboundary flux of nitrogen oxides at the 1987 level, while the effect-based 1999 Gothenburg Protocol sets fixed emission ceilings

10699

for the year 2010 (UNECE, 2004). The EU National Emission Ceilings (NEC) Directive (EC, 2001) defines slightly more ambitious 2010 emission ceilings for some of the Member States than the Gothenburg Protocol. The reason for this is possibly that the NEC was designed to deliver slightly different environmental objectives compared to Gothenburg Protocol in terms of ecosystem protection. The European Commission has also issued a number of Directives and instruments aiming to control NO_x emissions from specific sectors. These are principally the Large Combustion Plant Directive (Directives 88/609/EEC and 2001/80/EC), emission limits for engines used in non-road mobile machinery (Directive 97/68/EC), the Waste Incineration Directive (Directive 2000/76/EC) and the Euro standards for road vehicles (Directive 70/220/EC and revisions). The Euro standards for road transport emissions in Europe define the maximum limits of exhaust emissions from new vehicles sold in EU member states, but do not apply to vehicles already on the roads. No use of specific technologies is mandated to meet the standards, though available technology is considered in the policy development. Different Euro limits have been implemented between 1992 and 2005, and standards to be applied in 2010 have already been agreed.

Emissions from road transport have been determining NO_x emission levels for decades. Engine-out NO_x emissions consist mainly of NO (90–95%). NO is mainly formed by two mechanisms, basically the thermal (Zeldovich) and the prompt (Fenimore) mechanisms. The thermal mechanism is activated above 1600°C and is responsible for more than 90% of emissions from road transport. Reis et al. (2000) showed that road traffic may contribute substantially to exceedances of ozone indicators for both health and forests in Europe. Further, Carslaw et al. (2007) demonstrated the risk for the EU hourly limit of nitrogen dioxide ($200 \mu\text{g}/\text{m}^3$) not to be met by 2010 in European cities due to the recent developments in road transport. Globally, road transport is responsible for substantial increase in the concentration of tropospheric ozone (5–15%) not only in the vicinity of the source but also in remote areas (Granier and Brasseur, 2003; Matthes, 2007).

This paper documents how European anthropogenic road traffic emissions have

10700

evolved since the 1880s and investigates to what extent the decrease in emissions after 1990 can be linked to policy regulations. Our analysis links NO_x emission trends in Europe to the evolution of fuel consumption as well as to the changes in vehicle technology. It further distinguishes between the Eastern and Western European regions, where differences in the level of penetration of policy measures have an impact on the evolution of the emissions. Although the analysis covers a 125 years time span, the main focus is on the last 15 years, when European NO_x emissions have begun to decrease. The methodology developed is presented in Sect. 2 which also documents the data sources used in the analysis. Data quality is discussed in Sect. 3. Results on European trends in NO_x emissions are presented in Sect. 4, and the discussion on the effectiveness of policy measures is given in Sect. 5. Finally, conclusions are summarized in Sect. 6. Emissions from international shipping at European waters are analyzed in a forthcoming paper (Jonson et al., in preparation) and are thus not included here. With respect to the terminology adopted, NO_x emission figures correspond to NO₂-equivalents, except in cases where primary NO₂ emissions are explicitly discussed.

2 Methodology and data used

A European NO_x emission inventory that spans over 125 years has been compiled here to provide further insight in the evolution of European road traffic emissions. The inventory relies on available information on 1) activity data, 2) emission factors, 3) abatement level, and 4) the level of policy penetration. Such information is largely variable from period to period and for the different European countries and areas, and determines to a large extent the accuracy of the final results. Concise information on the information sources and an evaluation of the uncertainty associated with each source is also included in the following.

10701

2.1 1880–1985: EURONOX inventory

We have estimated anthropogenic fossil fuel and residential wood combustion emission by European country and sector every 5 years since 1880. The underlying activity data corresponds to the European historical country borders in the time span considered, and we distinguish between three different periods: 1880–1915, 1920–1945 and 1950–1985. The emission estimation methodology differs only for the periods 1880–1945 and 1950–1985. An advantage of this study with respect to previous global estimates (e.g. van Aardenne et al., 2001; Schultz et al., 2007) is that we have applied emission factors which vary with time and by country.

Fuel consumption in the period 1880–1945 is calculated from energy and industry statistics collected by Mitchell (1981) and supplemented with information from the World Power Conferences (1948). According to international consumption and production statistics, fuel consumption in this period is dominated by solid fuel use. Detailed activity data and reliable emission factors are not available for this period, thus we derive the emissions by scaling 1950 emission sectors per country backwards in time, based on the solid and liquid fuel consumption. We underline that one main implication is that possible important changes in the average emission factor for coal combustion during this period are not considered in our emission estimates.

In addition to the emissions from fossil fuel combustion, we also estimated biomass emissions in the residential sector. The coverage of wood consumption per country is rather incomplete in international statistics, particularly before 1950. In addition, we considered that underreporting of wood consumption by countries could be an issue. Therefore we made two different estimates, one based on an extrapolation of OECD activity data (OECD, 2004), and a second one assuming that each person living in rural areas consumes 1 m³ wood per year, with a conversion factor of 440 kg wood/m³ (Lunnan et al., 1991) and a historical rural population dataset (Marti-Henneberg and Tapiador, 2008). For both sets of activity data we used an emission factor of 100 g NO₂/GJ (IPCC, 2003) equivalent to 1.9 g/kg wood, using a heating value of 19 GJ/ton for wood

10702

(Sternhufvud et al., 2004). The results from this evaluation are addressed in Sect. 4.

With regard to the period 1950–1985, we distinguished between the originally OECD and non-OECD countries, based on the availability of activity data. A detailed breakdown of activity data is published in the OECD Energy Statistics (OECD, 1966, 2004),
5 and we estimated NO_x emissions from about thirty different sub-sectors aggregated per SNAP sector (Energy Production from Power-plants-S1, Residential-S2, Industry-S3 and S4, Road transport-S7 and Off-road machinery-S8). For the non-OECD countries we used the production figures of electric power in thermal power plants from the UN Energy Statistics (UNECE, 1976, 1980 and 1981), to deduce the amounts of lignite
10 and other fuels used in electric power plants. For most countries it has been assumed that mainly lignite was used in electric power plants. However, hard coal needs also to be included in energy budget of Poland and Hungary, while the Former Soviet Union (USSR) and Romania have been also using coal and natural gas, according to the statistics.

15 Coke production figures were used to deduce the amount of coal used for coke. The remaining coal was distributed between the industrial and the residential sector. For the non-OECD countries, gasoline was assumed to be consumed only in cars, or in other internal combustion engines. It was also assumed that gas oils must have been too expensive to be used except in internal combustion engines of cars, trucks, off-road
20 equipment and machinery, and in agriculture. In general, these uses are assumed to account for 80% of the gas oil consumption.

The emission factors used for this period are shown in Table 1 and are broadly based on the work by Pacyna et al. (1991), reviewing a large selection of country specific emission factors from national and international programmes, with a special attention
25 on Eastern Europe. The emission factors from Pacyna (1991) are representative of 1985. We have altered these emission factors to reflect changes over time and between countries. Further improvements have been carried out for emissions in the transport sector which is the main focus in our study. These improved emission factors are mainly based on the work by Samaras and Zierock (1996). Finally, in order to facil-

10703

itate crude conversion of the emission factors used in this study to other units, we have also indicated average calorific values in brackets in Table 1.

One main point is that the emission factors for motor vehicles increase with time during the period 1950 to 1985. This is to reflect the fact that the development of new
5 gasoline engines over this period led to less fuel-enriched mixtures, lower scavenging losses, and higher compression rates to improve fuel efficiency and to control CO and HC emissions that were the focus at the time (UNECE, 1958). As a side-effect of improved combustion, engines led to increased (thermal) NO_x emissions to the atmosphere. According to Samaras and Zierock (1996), emission factors of 20.4 g/kg were
10 applicable for gasoline cars without emission controls, i.e. vehicles produced before 1970 (Pre ECE R-15) but for vehicles with non-catalyst control (i.e. improved combustion) the emission factors increased to 36.7 g/kg (Table 1 footnote 3).

An increase in emission factors also occurred for diesel engines during this period, according to the US-AP42 (US EPA, 1991). The low emission factors for diesel engines are typical of engines with indirect injection. This design is not favoured for
15 modern, large trucks, which have direct-injection engines and higher compression ratios. The effective compression ratio may be further increased by turbo-charging, which further promotes the formation of NO_x. Emissions from for heavy duty vehicles (HDV) were not regulated until 1988 with the introduction of the ECE 49 Regulation. The
20 lower limit of the emission factor range for HDVs in Table 1 (30 g/kg) is comparable to Conventional HDVs included in COPERT, when these are converted per fuel mass used, (<http://lat.eng.auth.gr/copert>) and, further, to the uncontrolled NO_x emission factors in the GAINS database (<http://www.iiasa.ac.at/web-apps/apd/gains>) developed at IIASA. Measurement studies more often concern US and more recent vehicle fleets
25 (e.g. Kirchstetter et al., 1999; Yanowitz et al., 2000; Kristensson et al., 2003; Schmid et al., 2000; Kohler et al., 2004), but Ekström et al. (2004) report on-road optical remote sensing measurements in Sweden per vehicle technology class which support the upper limits of the emission factor ranges both for gasoline passenger cars and heavy duty vehicles in Table 1.

10704

The emission factors we have assigned to HDVs are lower in Eastern European (24–40 g/kg) than Western European countries (30–50 g/kg), to reflect the differences in vehicle technologies following the implementation of the ECE-R15 regulations in Western Europe. In addition, some Eastern European countries used to have a high proportion of 2-stroke engines which resulted in even lower average NO_x emission factors. In the extreme case of Former East Germany, more than 50% of the vehicles used to have 2-stroke engines, and the resulting emission factor for gasoline cars is consequently estimated at a much lower value (6–10 g/kg) than for the other countries included in this study (20–30 g/kg). While these simple considerations do not provide detailed and accurate inventories for each country, they may still give useful estimates of regional and temporal trends.

For stationary sources we do not include any variation of emission factors with time. We assign country specific emission factors for brown coal in thermoelectric power plants and in the industry according to fuel quality and combustion technology in the respective countries (UNECE, 1981; McInnes, 1996). This implies that emission factors for power plants are generally higher in Eastern Europe compared to Western Europe. The highest emission factor is assigned to Former Czechoslovakia (8 g/kg), followed by Albania, Bulgaria, Former USSR and Yugoslavia (7 g/kg), Poland (6 g/kg), Former East Germany, Hungary, Romania, Austria, Denmark and France (5 g/kg), Spain (4 g/kg), former West Germany Italy, Portugal and Turkey (3 g/kg) and Greece (2 g/kg). Due to lack of information about differences between countries in the industry sector, we have applied a uniform emission factor of 3 g/kg for the industry sector in all Eastern European countries. Emission factors for Western European industries are about 1 g/kg lower than those listed above for power plants.

It is worth noting that we do not include the gas associated with the production of coke from coal, hence available for combustion (e.g. coke oven gas or blast furnace gas from the iron and steel industry) in separate sectors. It is instead included in the emission factor for coke production itself, and this is why we apply an emission factor for coke production orders of magnitude larger than Pacyna et al. (1991). Combustion

10705

in the residential sector is assumed to occur mainly in small domestic boilers, and the emission factor chosen for combustion in oil refineries is taken from Takacs et al. (2004).

Emissions from international shipping and aviation are not included in this study. Further we do not include emissions not directly related to fuel consumption like nitric acid and fertilizer production. According to Pacyna et al. (1991), these are minor sources (0.5% contribution around 1980), as is agricultural burning of straw and stubble (less than 1%). Emissions from waste, which, according to data reported to the LRTAP Convention, contribute less than 1% to the national total in the 1980s is not included. No attempt has been made to include NO from soils although some authors (e.g. Stohl et al., 1996) argue that the emissions are mainly from arable land and should be therefore considered as anthropogenic.

2.2 1980–2005: EMEP NO_x inventory

For data on NO_x emissions after 1980 this study relies mainly on data from the EMEP (Cooperative Programme for Monitoring and Evaluation of the Long-range Transmission of Air Pollutants in Europe) programme. The EMEP inventory consists as far as possible of official data reported annually by 51 Parties to the Convention on Long-range Transboundary Air Pollution (LRTAP). The emission data is compiled at national level in accordance with the UNECE Emission Reporting Guidelines (UNECE, 2003) and the EMEP/CORINAIR Guidebook (<http://reports.eea.europa.eu/EMEP-CORINAIR4/en>). The national emission estimates are accompanied by an Informative Inventory Report (IIR) documenting the uncertainties in the data used and possible deviations from the recommended methodologies in the Guidebook. These emissions are annually reviewed and evaluated, to check for errors and identify areas where improvements may be necessary (e.g. Vestreng et al., 2007a). Since the adoption of the new reporting emission reporting Guidelines in 2002, the official reporting of data for the road transport sector includes valuable information per vehicle type i.e. Passenger cars, Light-duty vehicles, Heavy-duty vehicles, and Mopeds and

10706

motorcycles. Emission data available at this level of detail is mainly restricted to the period from 1990 onwards. Several countries report data on a more aggregated level up to 2000, i.e. total Road Transport (SNAP 7). The lack of detailed emission data in the 1980s and partly in the 1990s, has limited parts of our analysis to the aggregated SNAP 7 level. In addition to emission data, national reporting includes activity data for the historical years 1990, 1995, 2000 and 2005. These official activity data reported by countries have been used for the study of emission trends in the period 1990–2005. In the absence of reported data, our analysis for the period 1990–2005 relies on trends in fuel consumption and implied emission factors from the GAINS database. Historic fuel consumption data in GAINS are extracted from national and international energy statistics. For the period 1980 to 1990, we include fuel consumption data from the sources outlined above for the EURONOX inventory.

Table 2 presents national NO_x emission trends and gives an overview of the completeness of official emissions in the EMEP inventory between 1980 and 2005. The relative share of emissions from road transport (in brackets) is also listed. The emissions are based on officially reported data, to the extent possible. Countries which passed the EMEP review are highlighted with grey background in Table 2; a total of nineteen countries. The table identifies also a second group of countries for which reported data had to be completed by interpolation and extrapolation in order to achieve full emission trends for the period. These twelve countries are marked in bold italics. For the remaining countries, emissions were derived from other sources. The main source for non-official emission estimates in the EMEP inventory is data from the RAINS/GAINS model (<http://www.iiasa.ac.at/web-apps/apd/gains>) developed at IIASA. These emissions are not completely independent from those officially reported in that IIASA, through bi-lateral consultations, may include data provided by the countries themselves. The RAINS/GAINS model is now capable of reproducing national emissions for NO_x for almost all Parties with an uncertainty margin of less than 5% (UNECE, 2007). Because there are a few countries for which neither official nor RAINS/GAINS data are available, EDGAR emission data (<http://www.mnp.nl/edgar>) are also included.

10707

The EDGAR inventory is a global inventory but it does not have the same level of detailed vehicle classification as the RAINS/GAINS model. There are nine countries for which RAINS data have been used and these are marked with a star in Table 2. The emission estimates for the remaining four countries rely on EDGAR emission data. The coverage of reported emissions is about 40% in the 1980s, increasing to nearly 60% after 1990. The level of confidence is considered to be higher for the reported and reviewed emission data, due to country specific insight and the detailed input to the calculations.

3 Data quality

As indicated from the discussion above, the level of accuracy of the data used all through the NO_x inventory from 1880 to 2005 increases as we approach recent times. In this section we document the uncertainties in the EMEP and EURONOX inventories, and justify the merging of these two inventories.

3.1 Uncertainties in the EMEP inventory

There are recognised uncertainties in the selection of emission factors and even though national statistics of activity data as compiled from e.g. data reported by individual facilities, registration offices and different surveys are in most cases reliable, there is also an element of uncertainty in this basic input to the national emission calculations. Discrepancies between actual and apparent national emission estimates are also introduced when emission data are reported, in line with the reporting Guidelines, based on fuel sold rather than fuel used. This is because the amount of fuel sold in a country may be strongly influenced by “fuel tourism”. This is a term used for retail purchase of fuel in one country for consumption abroad, mainly due to fuel price differences. Although emissions associated to the combustion of this fuel do not occur in the country where the fuel was sold, the UNECE reporting Guidelines require Parties to report

10708

these emissions as national emissions. The effect of this fuel tourism is shown to have opposite and equally large effect for countries with high “green taxes” as discussed in Sect. 5 in the case of Germany. The implication is that while the European emission trend for NO_x may be correctly reflected, the national (road transport) emission trends for several European countries may be affected by the tax and transit levels.

The uncertainty level for national inventories included in the EMEP inventory is based on information given in the Informative Inventory Reports (IIR) accompanying the official submissions emissions under the LRTAP Convention. Based on a review of this information from a limited number of countries, the uncertainty in national emissions is considered to be between 8% and 23% for Western Europe and around 25% for Eastern Europe. The EMEP inventory contains in addition emission estimates from the RAINS/GAINS model, and according to Schöpp et al. (2005), the uncertainty in these emissions are comparable to those reported by the countries. Uncertainty estimates for individual sectors are not reported by the Parties, but Schöpp et al. (2005) indicate that the sector uncertainty is higher, and might be nearly three times larger for emissions from gasoline passenger cars and diesel heavy duty trucks. Kuhlwein and Friedrich (2000) estimate the statistical error in transport NO_x emissions in West Germany to be 16–22%, comparable to the results for United Kingdom estimated by Schöpp et al. (2005). The above uncertainty ranges are applicable from 1990 onwards (Vestreng et al., 2006). Quantitative uncertainty estimates for the 1980s are not available, but they are likely to be larger, due to the lower coverage of reported emissions and absence of published non-official estimates. In addition, recalculation of emission data by many Parties are only performed from 1990 onwards, hence the accuracy in the 1980s emissions may not benefit from methodological improvements in emission estimation.

A complementary way to assess the validity of emission data is to combine model and observation data. The general downward trend in EMEP emission data from 1990 onwards have been confirmed by a recent model study by Jonson et al. (2006). The study concludes that even though the EMEP model tends to overpredict winter concen-

10709

trations and underpredict summer concentrations compared to measurements, NO₂ levels and seasonal patterns are well captured. Further, Fagerli and Aas (2008) show that the reduction in EMEP NO_x emissions between 1990 and 2003 are comparable to the downward trend in measurements of nitrate in precipitation.

A trend study by Konovalov et al. (2008) applying inversion techniques with GOME and SCIAMACY measurements between 1996 and 2004, broadly confirms that the NO_x emission trends in Europe have been decreasing, and further indicates that the quality of the EMEP inventory has increased over the last few years. Our evaluation of regional differences in inventory uncertainties is in agreement with the above study, where particularly large differences between the EMEP and satellite data are found in Balkan countries, Georgia, Russia and Turkey. It is however important to consider that there are limitations both in the model and in the observations when such top-down methodologies are applied to derive emission trends, and we suspect that the lack of reduction in emissions seen for Italy in Konovalov et al. (2008) could also be due to limitations in the vertical extent of the chemical transport model applied for the calculations.

3.2 Uncertainties in the EURONOX inventory

The quality of the EURONOX inventory is much influenced by availability and accuracy of the activity data. There are some gaps in the statistical activity data, but in general the quality of the emissions is better from 1950 onwards than before 1950, when less detailed data was available and emissions had to be extrapolated backwards in time. Also, the uncertainty is higher for Eastern than for Western European countries, due to the lack of activity data in specific consumption sectors. There are also uncertainties linked to the selection of emission factors for different sectors and the choice of method, e.g. more generalized emission factors. Emission measurements from stationary sources have only become available in the last 30 years, while measurements of mobile sources came even later. The quality of the emission estimates is hence expected to increase as we approach recent times.

10710

The comparability between the EURONOX and EMEP inventories has been assessed for the two common years, 1980 and 1985. Table 3 shows differences in national total and road transport emissions per country in both inventories. The agreement on the national total levels is generally good, with an underestimation of the European total in EURONOX relative to EMEP by 8% and 6% in 1980 and 1985, respectively. The better agreement in 1985 is probably because the emission factors applied are more representative for 1985 than for 1980. The general overestimation indicates that the emission factors do not reflect any of the emission controls already effective in the 1980s. Particularly large differences between reported and estimated emissions occur for Czechoslovakia, where we apparently underestimated the emissions by a factor 2. This occurs in spite of our use of emission factors which take into account both the very high energy content of Bohemian lignite and the combustion technology in the Czechoslovakian power plants. If the reported Czechoslovakian NO₂ emissions are correct, the NO₂/capita ratio in 1980 would have been 74 kg/capita, i.e. far outside the NO₂/capita range of 20–45 kg elsewhere in Europe, and suggests that the sum of the reported Czech and Slovakian NO₂ emissions are too high. Kopáček and Veslý (2005) have estimated the Czechoslovakian emission to about 710 Gg, thus is more of support of our calculations. The differences in emission estimates for Sweden are also quite large (factor 1.5). The use of gas oils in off-road machinery could explain the underestimation relative to the official emission in this case. Such large differences will be discussed with the national emission experts during the forthcoming reviews.

The comparability is lower when sector data are considered, but the differences are still mostly within the uncertainty range indicated by Schöpp et al. (2005). In contrast to the national total emissions, road transport is generally increasingly overestimated by the EURONOX inventory, indicating that the emission factors applied might have been too high for some countries. The differences are however particularly large for some of the Eastern European countries, and here the discrepancies in sector emissions could also be attributed to the lack of detailed activity data statistics. While it is clear that

10711

much more detailed information about the conditions in each country would have been desirable when developing the EURONOX inventory, the agreement with the EMEP data is considered sufficient to merge the two inventories in 1980, by scaling the EURONOX inventory to the relevant EMEP sectors. In order to account for the sources not included in the EURONOX inventory, we scaled the residential sector also together with the EMEP agricultural and waste emissions.

3.3 Comparison with other estimates

The combined EURONOX and EMEP inventory is compared to independent inventories both at national (anthropogenic) total and at road-transport levels. The preferred option would have been to make the comparison by country, but the other inventories were not available in such detail. Schöpp et al. (2003) has compiled a NO_x inventory based on a study by Dignon and Hameed (1989) between 1880 and 1960. The Dignon and Hameed (1989) inventory is merged with estimates from an old version of the RAINS model from 1960 onwards. The European NO_x trend presented in Schöpp et al. (2003) differs considerably from our work in that emissions are consistently higher in Schöpp et al. (2003) over the whole 1880–2005. The difference between the inventories is particularly large in the 1950s, and amount to nearly 40% in 1960 at the European level. Dignon and Hameed (1989) derive emissions by regression analysis from total fuel consumption. It is likely that our refined approach, with application of representative emission factors in distinct fuel consumption sectors, is the main reason for the large discrepancy between these two inventories.

A global inventory published by van Aardenne et al. (2001) is available for the years 1890 to 1990 in ten-year intervals. The road-transport emissions are not separately documented but are included in the fossil fuel combustion sector. National and road-transport data per decade between 1960 and 2000 were made available to us on a regional level for the more recent RETRO inventory (Schultz et al., 2007). EDGAR data (<http://www.mnp.nl/edgar>) are available per country and sector in five-year intervals between 1990 and 2000. Comparison with these three inventories has been

10712

made on the regional “OECD Europe” and “East Europe” level, as defined by EDGAR (<http://www.mnp.nl/edgar>). The comparison is made from 1920 onwards as such regional comparisons are hampered by differences in country borders, particularly before 1920.

5 The OECD emissions presented in Fig. 1 represent between 97% (1920) and 55% (2005) of the total European emissions according to our inventory. Our work and the van Aardenne et al. (2001) study compare well both in terms of trend and national emissions level over the whole hundred year time span (Fig. 1). Van Aardenne et al. (2001) emissions are slightly lower (less than 20%) compared to our work in 1970 and 1980.
10 Since detailed activity data is available for this period, the difference is apparently due to lower emission factors applied in the van Aardenne et al. (2001) inventory. The RETRO inventory defines the peak in total emissions in 1980, contrasting both our work and the work by van Aardenne et al. (2001). The RETRO national total estimates are lower than this study, the van Aardenne et al. (2001) and the EDGAR inventories for
15 all years but 2000, where the inventories coincide. The underestimation in the RETRO inventory compared to EMEP can only be partly explained by the incompleteness in the RETRO inventory with respect to national navigation, railway, waste treatment and disposal, and cement manufacturing. The difference in trend between the RETRO and the other inventories between 1980 and 2000 seems to be due to application of more
20 efficient abatement in stationary sources, as the trend in road transport compares fairly well both by our work and the EDGAR inventory. Based on these considerations, we conclude that our work is closer to the van Aardenne et al. (2001) and EDGAR estimates than the RETRO emissions at the total OECD level, even though the RETRO inventory for the road transport sector are similar to ours.

25 The much larger relative differences in emission level (more than 100% in some years) and trends for both total and road transport emissions in “East Europe” confirm that the uncertainties are larger in this area (Fig. 2). The comparison indicates an overestimation of emission totals in our study between 1980 and 1990 comparable to the anticipated too high reported emissions from the Czech Republic and Slovakia previ-

10713

ously discussed. Road transport emissions increase in the EDGAR inventory between 1995 and 2000, contrasting the EMEP emissions. This increase is reflected in the totals, and results in an overestimation of the EDGAR emissions in year 2000 compared both to our study and to the RETRO emissions. The increase in transport emissions in
5 EDGAR follows the trend in fuel consumption in this area, and does not seem to take into account that emissions have decreased in line with the implementation of Euro standards in countries like Poland and the Czech Republic as discussed below. The RETRO road transport trend is much weaker than in our work, indicating that emission factors vary less with time.

10 **4 Results on European emission trends 1880–2005**

4.1 European total trends

Figure 3 shows the trends in solid and liquid fuel consumption from 1880 to 2005 as compiled for this study. The total fuel consumption increased by more than a factor of ten over a period of a hundred years (1880–1980). Before 1950, solid fuel was the main
15 energy carrier in Europe, and the consumption increased steadily from 1880 onwards; the increase only interrupted by the economic depressions in the 1930s and later during the Second World War. Liquid fuel consumption showed a dramatic increase after 1950, among other reasons due to the availability of oil imported from the Middle East. The results presented here trace the relative importance of liquid fuel consumption in
20 comparison with solid fuel use. Between 1950 and 1970 the consumption of liquid fuel increased by a factor 18, and has exceeded the solid fuel consumption in all years since 1970. While European solid fuel consumption continued to increase until to the end of the 1980s, the increase in liquid fuel consumption ceased between 1970 and 1980, decreased thereafter until about 2000, and then increased again. The stabilisation and
25 decrease in liquid fuel consumption after the 1970s is a result of the high oil prices following the oil crises (e.g. Glover and Behrens, 2006) and is also due to decreased

10714

consumption in Eastern European countries. Solid fuel consumption drops sharply between 1990 and 2000 and increases thereafter. The decline in solid fuel is mainly due to decrease in hard coal all over Europe. In addition the consumption of brown coal went down in the EU area.

5 The trends in European NO_x emission related to these fuel consumption results are presented in Fig. 4, where also trends per sector are included. The sectors included in this analysis broadly follow the SNAP categorization, i.e. road transport, energy production from power plants, industry, off-road and residential. NO_x emissions from residential wood combustion (not shown separately in Fig. 4) only contribute in appreciable amounts to the total NO_x emissions around the turn of the 19th century when the contribution is estimated to be around 14%. From 1900 and onwards emissions from biomass have been minor (1–9%). Our results indicate that the residential wood consumption statistics are reasonable compared to our “rule of thumb” estimate.

10 National shipping and domestic aviation is included in the off-road sector. Agriculture and waste, being in general minor NO_x sources, have been merged with the residential sector. This distinction of sectors clearly shows the dominant effect of road transport in emissions over the last 35 years. Based on the developments in road transport, we have distinguished five emission trend regimes between 1880 and 2005. In the first regime, 1880–1950, the historical total NO₂ emission trend follows the moderately growing fuel consumption. Between 1950 and 1980 (the second regime), NO_x emissions grew steeply by a factor of 4.4, i.e. almost twice as fast as the sulphur emission increase during this same period (Vestreng et al., 2007b). The NO_x emission trend was strongly related to the increase in road transport emissions during this period, as indicated in Fig. 4. Already in 1970 the road transport emissions became the single most important source of NO_x with a share of nearly 30% of total emissions. The growth in the second and third largest sources (power plants and industry) was considerably less. The large change in the residential and off-road sectors between 1970 and 1975 shown in Fig. 4 was due to a reduction in the domestic consumption of residual fuel oil and an increase in diesel consumption in the agricultural sector. While we find the

10715

decrease in residual oil for heating plausible, we suspect that the detailed statistics we have used on diesel consumption prior to 1970 might be defective.

15 In the third regime, 1980–1990, the share of NO_x emissions from road transport is large (about 40%), and has remained relatively constant at the European level for the last 25 years (Table 2 and Fig. 4). Total NO_x emissions peaked in 1990, partly due to continued increase in road transportation activity up to this point in time, and partly to the fact that emissions from stationary sources remained relatively stable between 1980 and 1990. The fourth regime, 1990–2000, is characterized by a steep decline in NO_x emissions. The highest share of road transport to the total emissions (42%) is found around year 2000, and does not coincide with the peak in total NO_x emissions. This is due to the slower reduction rate of road-transport emissions (22%) relative to emissions from power plants (42%) and the industry (33%), between 1990 and 2000. As a result of the combined reductions, the total NO_x emissions monotonically decreased by 32% between 1990 and 2005. The largest reductions took place in the first half of the 1990s. The reasons of this decline are different in different parts of Europe and will be explained in the next section. Finally, in the fifth emission trend regime, 2000–2005, the downward emission trend has flattened out.

4.2 Trend differences between European countries in the last twenty-five years

20 We focus our analysis on the last three emission trend regimes, i.e. 1980–2005 for two main reasons. First, large changes in the emission trends can at least partly be associated to the technological development and policy regulations in this period. Second, the emission data uncertainty should be lower than in the period before 1980, as indicated in Sect. 3, and this may lead to more solid conclusions. The analysis particularly addresses road-transport, which is the most significant sector and a number of policy regulations have been developed to abate NO_x emissions from vehicles. The effectiveness of these regulations in Eastern and Western European countries is separately assessed.

The country specific details in NO_x emission trends after 1980 for both national

10716

and road transport emissions (percentage contribution in brackets) are highlighted in Table 2. The largest contributors are the Russian Federation, United Kingdom and Germany. The total NO_x emissions in Europe increased by 5% from 1980 to 1990 due to increased emissions in most countries but a few notable exceptions like Germany and France where emissions from power plants and the industry were reduced.

NO_x emissions decreased in most countries between 1990 and 2005, but there are substantial differences in the emission trends depending on the socio-economic and political situation in each country. A large reduction appears between 1990 and 1995 (Fig. 4) due to the disintegration of the Soviet Union in 1991. As a result of the economic recession, the reduction in NO_x emissions from the power plants and the industry was twice as large in the east as in the west, despite the introduction of specific abatement measures in the latter.

Except for the effect of this outstanding political situation, the NO_x emission trend over the period 1990 to 2005 has been dominated by changes in road transport. However, there are large differences between the east and the west. In Western Europe, road transport has been the dominant NO_x emission source over the whole period 1980 to 2005, while power plants were the most important NO_x source in Eastern Europe until 1995. For example, the road transport contribution in 1990 varied from less than 20% of total NO_x in Ukraine and Kazakhstan to about 60% in France and Switzerland. As a result, 70% of the total European road transport NO_x emissions in 1990 came from Western Europe. In 2005, this share dropped to 63% mainly due to reductions in Western European emissions, but also due to increase in emissions in the recovering economies in Eastern Europe.

Due to the dominant character of road transport emissions, Fig. 5 presents the trends in road transport NO_x emission, separately for the periods 1980–1990, 1990–2000 and 2000–2005 together with a reference map of emissions in 2005 (upper left map). The legend accompanying the difference maps (–20% to +20%) has been chosen to highlight the main differences, but the percentage differences might in certain cases exceed ±50% in any of the three periods considered.

10717

4.2.1 1980–1990

Road transport emissions in Europe increased by 13% in the period 1980–1990 despite a 10% reduction in Eastern Europe (cold colours in the upper right map of Fig. 5). The reduction in the east is linked to decreased fuel consumption due to income deterioration, as a consequence of the inefficiency in resource allocation (investments) (Gros and Steinherr, 1991). Exceptions to this general picture, where emissions increased, are Armenia, the Republic of Moldova, Hungary, Slovenia, Croatia and Estonia. While the explanation for the increased emissions in Hungary can readily be linked to the increase in gasoline consumption, the situation is not clear for the Former Yugoslav and USSR Republics. Fuel consumption data for individual Former Yugoslav and USSR Republics have not been available to us for the period 1980–1990, thus firm conclusions regarding the reasons behind the apparent increase in emissions cannot be drawn. Based on the rather stable fuel consumption trend in both the Former USSR and Yugoslavia between 1980 and 1990, it is not unlikely that fuel consumption increased in some of these Republics and decreased in other parts of this region.

In most of Western Europe and Turkey, road transport emissions increased between 1980 and 1990 (warm colours in the upper right map of Fig. 5). The overall increase was 27% in Western Europe. Fuel consumption went down or stabilized also in this region due to the high oil prices following the oil crisis in the 1970s. At the same time, early non-catalyst controls introduced with the different steps of UNECE Regulation No. 15 (1970–1983) were associated with an increase in NO_x emissions from vehicles (Berg, 2003). Due to relatively slow fleet turnover, as further discussed in the next section, the introduction of the ECE-R15 regulation maybe responsible for the overall increase between 1980 and 1990. In some Western European countries though, road transport emissions decreased between 1980 and 1990. These are Sweden, Belgium, Luxembourg, Austria, Switzerland, Cyprus and Malta. The fuel consumption went down also in these counties. A possible explanation for the emission decrease could be the early introduction of diesel passenger cars. In the case of Turkey, the

10718

increased emissions are due to a substantial (80%) increase in gasoline consumption.

4.2.2 1990–2000

In the period 1990–2000, road transport emissions decreased by 23% in Europe, and reductions were evident both in the east and the west (Fig. 5, lower left map). In Eastern Europe, the decrease in emissions is associated with a decrease in fuel consumption in former Soviet republics, Romania and Bulgaria. A country's transport volume is closely linked to its GDP, and the overall decrease in road transport is an effect of the restructuring of the economies after the disruption of the Soviet Union in 1991. Russia is an important trade partner, so the depression also affected countries outside the Union. Further, the infrastructure in this region which was already rather poor further decayed during this period (EEA, 2007). Decreased emissions in other Eastern European countries are linked to decreased emission factors, rather than decreased fuel consumption. The share of the high-polluting car fleet built in Eastern Europe decreased in these areas between 1990 and 2000 as the increase in the stock of vehicles is due to imports of cleaner cars from Western Europe. This development took place also in the Czech Republic, but here the increased share of lower NO_x emitting cars only damped the increase in emissions. Albania and The Former Yugoslav Republic of Macedonia increase their emissions due to increased fuel consumption, without an accompanying decrease in emission factors. The 7% decrease in road transport emissions reported by Croatia cannot be explained without assuming a decrease in emission factors, as both GAINS and IEA report increased fuel consumption in the transportation sector in Croatia between 1990 and 2000.

In Western Europe, the introduction of improved vehicle technologies and stringent inspection systems related to the Euro standards has been the primary force in reducing NO_x road traffic emissions in the period 1990–2000, despite economic growth and increases in fuel consumption. All countries but Portugal, Spain, Greece, Turkey, Cyprus, Malta, Austria, Ireland and Luxembourg reduced their emissions (Fig. 5 lower left map). These nine countries which increased emissions between 1990 and 2000

10719

can be divided in three groups based on the possible causes for the emission growth. The high age of the vehicle fleet combined with increasing number of vehicles may explain the lack of reductions in the first group, containing Portugal, Spain and Greece. In the second group with Turkey, Cyprus and Malta the main reason for emission increase is that the Euro standards were not applied at the same time as in the rest of Europe. Increase in emissions reported from Austria, Ireland and Luxembourg are caused by fuel tourism as defined in the previous sections. Austria and Ireland provide road transport emissions both according to fuel sold and fuel used. Their estimates for NO_x emissions calculated on the basis of fuel used show a decreasing trend between 1990 and 2000, opposing the data reported as requested by the UNECE Guidelines according to fuel sold. The reason for the increased emission in Austria is a large increase in emissions from heavy duty vehicles (Anderl et al., 2007). In Ireland, the reason is that fuel is less expensive in Ireland than in the United Kingdom during this period. UK fuel prices apply to Northern Ireland, thus drivers tank in Ireland (DEHLG, 2006).

4.2.3 2000–2005

In the period 2000–2005, road transport emissions in Europe continue to decrease by 11%, albeit less than in the preceding decade (23%). Fuel consumption in the traffic sector increased in all European countries except in Germany. In Germany high tax on fuel combined with improvements in vehicle technology, result in a considerable decline in diesel consumption as further discussed in Sect. 5. The situation from the 1980s (Fig. 5 upper right map) with decreasing emissions in the east and increased emissions in west is reversed in this period (Fig. 5 lower right map).

Increase in emissions from Eastern Europe follows the increase in fuel consumption (Fig. 5 lower right map). The recovering of the economy is responsible for the emission growth, and it is illustrative that loans for transport from the European Bank for Reconstruction and Development to the EECCA countries have mostly financed roads after year 2000. This contrasts with the previous periods when rail and port projects

10720

dominated (EEA, 2007). The EECCA countries have their own car industry, so new western technologies will not necessarily become standard. Another reason why emissions in EECCA countries increase may be related to use of lead as an additive to the fuel. Lead additives destroy the catalysts, and are not completely abandoned due both to lack of regulations and to a claimed black market for leaded gasoline. In addition to this, the price of fuel is low and even subsidised in some countries (EEA, 2007). In Belarus, emission decreased between 2000 and 2005. There is no essential production of cars except for heavy duty vehicles here. Produced lorries comply with Euro 2 and later standards and passenger cars are imported. Import of cars which not comply with certain Euro standards are not directly prohibited, but the older cars are imposed higher tax. The situation with respect to how the introduction of Euro standards has influenced the emission trend is mixed for the EU-10 countries. While Hungary, Latvia, Lithuania and Slovakia report an increase in emissions between 2000 and 2005, due to less effective implementation of the Euro Standards, decrease in emissions are seen in Poland, Czech Republic, Estonia and Slovenia. In addition, Croatia which according to the Belgrade report (EEA, 2007) implemented the Euro standards from year 2000 decreased their emissions.

Contrasting the general increase in Eastern European emissions, the decrease in emission continues in Western Europe between 2000 and 2005. The only countries where emissions increased were Turkey and Austria. In Turkey emissions increased because of lack of abatement measures and Austria due to fuel tourism.

5 Effectiveness of policy regulations in the transport sector

As indicated in Figs. 3 and 4, the European road transport emissions have been decoupled from the liquid fuel consumption since 1995. This section investigates to what extent the decrease in NO_x transport emissions can be associated to the introduction of the Euro standards for both passenger cars and heavy duty vehicles. By convention, these standards are denoted with Arabic numbers for passenger cars and light duty

10721

vehicles (e.g. Euro 1, 2, . . .) and Latin numbers for heavy-duty vehicles (e.g. Euro I, II, . . .). Table 4 shows the NO_x relevant emission standards per vehicle category and the associated emission control technology in the European Union after 1992. To facilitate comparison with reported implied emission factors, emission standards in PJ/Tg are also included.

The analysis here investigates to what extent emission factors calculated on the basis of officially reported road transport emissions and activity data (implied emission factors) comply with the Euro standards. This information is only available from 1990 to 2005 for ten Western European countries (Austria, Denmark, France, Germany, Netherlands, Norway, Portugal, Spain, Switzerland and United Kingdom) and can be retrieved from the EMEP database (<http://webdab.emep.int>). These ten countries represent more than 50% of total European emission from road transport in 1990, and they are considered to represent the situation in Western Europe. For Eastern Europe, relevant data are available for 2005 for seven countries (Estonia, Lithuania, Macedonia, Poland, Romania, Slovakia and Slovenia). Therefore, a separate analysis has also been undertaken for this particular year.

5.1 Trends in emissions and fuel consumption by country and vehicle class

The fuel types considered here are gasoline and diesel. The reported consumption of hydrogen is negligible and also the reported consumption of compressed natural gas (CNG) and liquefied natural gas (LNG) is very low to affect the trends. In addition, the fuel consumption in mopeds and motorcycles is small compared to other vehicles categories and therefore these have not been considered in the following analysis. It is a limitation to our analysis that the reported emissions do not distinguish between gasoline and diesel use in vehicles. The implied emission factors are thus calculated based on total emissions and fuel consumption for each vehicle class. We argue that our conclusions are robust, albeit we cannot calculate IEFs separately for PCs and LDVs. The reason is that, although the reduction efficiency for NO_x in gasoline PC is quite high even for the first generation catalysts, there were relatively few catalysts

10722

installed in 1990. This implies that we can assume emission factors to be higher for gasoline than diesel PC over most of the period 1990 to 2005. Thus our combined IEFs can be considered lower than the IEFs for gasoline vehicles alone.

In Western Europe, the overall trend in fuel consumption shows that petrol consumption decreased (20%), while diesel consumption increased (90%) between 1990 and 2005. The shift to diesel is the impact of the European Automobile Manufacturers Association's commitment on the reduction of CO₂ emissions from passenger cars (Commission Recommendation 1999/125/EC) (ACEA, 2007). This agreement promoted the use of diesel passenger cars because they have up to 30% higher fuel efficiency than gasoline cars of similar size. The net fuel consumption in road transport increased about 23% from 1990 to 2005. Passenger cars consumed by far the largest share of fuel (60%), followed by heavy duty vehicles (28%) and LDVs (12%). The promotion of diesel cars via the ACEA Commitment greatly benefited the curtailment of greenhouse gases. At the same time, it should not be forgotten that diesel passenger cars emit as much as three times higher NO_x emissions per kilometre than gasoline cars of the same emission standard. Just to put it into perspective, assuming that the increase in fuel consumption would have originated from increase in petrol rather than diesel consumption (thus diesel consumption remaining at the 1990 levels), this would have led to some 1/3 lower NO_x emissions in 2005.

With respect to road transport emissions of NO_x, these decreased by 44% between 1990 and 2005 in Western Europe, despite the increase in fuel consumption. The emission reductions were largest for PC (63%) followed by HDV (21%) and LDV (2%). Figure 6 compares the trends in total fuel consumption from 1990 to 2005 with individual trends for gasoline and diesel consumption and with the total NO_x emissions for passenger cars in selected Western European countries. NO_x PC emissions decrease monotonically while diesel consumption substantially increases (by nearly 200%) and gasoline consumption moderately decreases (by 19%). There is a clear decoupling of emission and fuel consumption of passenger cars already since 1990, as result of the developments in vehicle emission control technologies.

10723

For HDV the situation is more complex. Fuel consumption increased in all countries between 1990 and 2005, except in Germany, where HDV consumption decreased by 30% between 2000 and 2005. This substantial decrease in diesel sold is not likely due to technological developments alone, but also due to the high tax on diesel in Germany. The high fuel prices in Germany prevent transit traffic refuelling, and promote fuel tourism to other neighbouring countries. Emissions from HDV between 1990 and 2005 decreased in all countries, except in Spain and Portugal and Austria, where emissions increased, by more than 200% in the case of Austria. Austria is a counter case to Germany, in that some 30% of the diesel sold is consumed outside the country. The onset of emission reduction from HDV comes almost ten years later than the corresponding turning point of passenger cars emission. The main reason for the delay in HDV emission reduction is the inefficiency of Euro II standards in addressing NO_x. With regard to LDVs, their fuel consumption increased in all countries. Their emission levels have remained relatively stable compared to the emission trends in PC and HDV, with slight increases or decreases in equally many countries. The above results show that the implementation of Euro standards has contributed to a decoupling of emissions and fuel consumption of all vehicle classes in Western Europe since 1990.

5.2 Trends in implied emission factors

We have derived implied emission factors (IEF) between 1990 and 2005 based directly on reviewed officially reported emissions and total (gasoline plus diesel) fuel consumption. In this way we can compare the average emission level of the whole fleet in each country, with the emission levels expected when developing the Euro standards. The results for Western Europe are presented in Fig. 7, which shows that the implied emission factors decrease for all vehicle classes from 1990 to 2005. The average IEF reductions for all countries examined in this period are 67%, 42% and 35% for PC, LDV, and HDV respectively. The periods with largest IEF reductions vary with vehicle class and country. For PC, the largest IEF reductions (35%) occurred between 1995 and 2000 while max reductions for HDVs (20%) appeared five years later (2000

10724

to 2005). On average, the IEF reductions from LDVs remained relatively constant, at 17%, in all five-year periods. We know today that the introduction of electronic controls in Euro II (1997), and less so in Euro III (2001) heavy duty engines led to excessive NO_x emissions over operation modes that were not included in the type-approval test (Hausberger and Rexeis, 2004). As a result, countries with a fast turnover of their HDV fleet were delayed in meeting the stringent emission standards expected. This is the reason that the mean HDV-fleet emission factors in several countries (UK, Netherlands, Austria, Denmark, Switzerland) in 2000 still appears higher than the Euro I emission standard introduced eight years before (1992). The situation improves in 2005 with only Austria and Denmark appearing to have HDV emission levels clearly beyond the emission standards 8 years ago (Euro II).

The conclusions related to the effectiveness of the Euro emission standards for passenger cars are less straightforward, since the IEF is a composite value of gasoline and diesel vehicle emission levels, while separate emission standards have been in place, depending on the fuel used. In general, Fig. 7 shows that the average fleet emission level in 2000 in several European countries corresponded to a level between the gasoline and diesel Euro 1 levels, eight years ago. Spain, with a rather old vehicle fleet, fails to meet the Euro 1 emission standard level even eight years after the introduction of the standard. Even in 2005, the average emissions of the Spanish PC fleet are only marginally below the diesel 1992 levels. On the other hand, average PC fleet emissions in Germany and Switzerland seem quickly (e.g. within five years) to attain the emission standards, despite the large fleet of diesel passenger cars. A similar decadal delay in effective implementation of the Euro standards is also seen for LDVs.

Due to lack of reported data, changes over time in IEF cannot be determined for Eastern European countries. The analysis for Eastern Europe is restricted to year 2005. Table 5 compares implied emission factors for PC, HDV and LDV in Eastern and Western European countries. IEFs in Eastern European countries are a factor of 2.3 and 1.4 higher than in Western European countries for PC and LDV respectively. This is because the Euro standards did not fully apply to Eastern European countries

10725

before their accession to the European Union in 2004. The result further implies that the implementation of technological measures to abate road transport emissions has been less effective in Eastern Europe, due to a slower turnover of vehicles towards more modern, less polluting technologies.

The conclusion from this analysis is that Euro standards have clearly facilitated a substantial reduction in the road transport emissions in Western Europe. There has however taken some eight to ten years for policy regulations to come fully in effect in several European countries. This is the effect of the slow turnover of the vehicle fleet in many countries but also due to the deviations between the emissions in real-world conditions, compared to the type-approval driving cycle. Life cycle assessments of car fabrication might shed light to whether or not the policy should increase the incitement to a faster vehicle turn-over.

6 Conclusions

The significant increase of liquid fuel consumption in Europe between 1950 and 1980 led to an unparalleled historic increase of NO_x emissions from road transport by a factor 14 (Fig. 3). Road transport emissions have been the main source of NO_x in Europe already since the 1970s and are currently responsible for about 40% of total anthropogenic emissions. Technological and policy developments to abate European emissions have clearly facilitated a substantial reduction in the NO_x levels. Between 1990 and 2005 emissions decreased by more than 30% (Fig. 4, Table 2), but have now started to increase in many Eastern European recovering economies.

Based on the development in road transport emissions, we determined five NO_x emission trend regimes in Europe. While the emission trends in the first two of these (1880–1950 and 1950–1980) are mainly determined by the development in fuel consumption, the last three regimes are driven more by policy developments. In the third regime (1980–1990) road transport emissions decreased in Eastern Europe and increased in Western Europe (Fig. 5 upper right map). These regional trends resulted

10726

in an overall increase in NO_x emissions. The emission decrease in Eastern Europe was linked to deterioration in incomes, followed by a decrease in fuel consumption. In Western Europe, fuel consumption increased despite historical high oil prices in the beginning of the period. In addition, the ECE-R15 regulations introduced in the 1970s to improve combustion in motor vehicles, increased NO_x emission factors and NO_x emissions peaked in 1990. In the fourth regime (1990–2000), large reductions in transport emissions took place all over Europe (Fig. 5 lower left map). In Eastern Europe, decrease in emissions is linked both to decline in fuel consumption in Former Soviet Republics, and to a reduced share of high NO_x emitting vehicles in other Eastern European countries. The energy consumption increased in Western Europe, but policy regulations fostered technological development and implementation of measures, which resulted in large decreases in road transport emissions. In the fifth regime (2000–2005), the emissions pattern from the 1980s was reversed (Fig. 5 lower right map). Emissions continued to decrease in Western Europe due to the implementation of stricter control measures. On the other hand, emissions increased in line with the GDP in large parts of Eastern Europe. Historically, the most striking difference in NO_x emission distribution between Eastern and Western Europe is the much higher contribution from road transport in Western Europe. However, as a result of the recent development in road transport emissions, the emission levels in Eastern and Western Europe are now rapidly approaching each other.

Environmental and political concerns drove the decision to abate NO_x emissions, resulting in the NO_x Protocol under the Convention on Long Range Transboundary Air pollution (UNECE, 2004) in 1988 and were then followed by the Multi-effect Protocol in 1999. In addition, European Commission regulations specifically targeting the transport sector (Euro 1–4) were introduced between 1992 and 2005. The UN Protocol obligations may have led to substantial reductions, but we found that it was a lot easier to trace the effectiveness of the sector specific regulations. This is because the stationary emissions closely followed the trend in solid fuel consumption, while we found that traffic emissions were decoupled from fuel consumption already since the 1990s.

10727

The emission limits for vehicle exhaust introduced by the Euro standards (Table 4) led to a substantial decrease in emission factors for all vehicle types in the EU region (Fig. 7). Despite a significant increase in the fuel consumption over the whole period 1990–2005, emissions monotonically decreased in the case of passenger cars (Fig. 6). There is clear evidence that the policy approach taken to reduce NO_x emissions has been effective in bringing NO_x levels down. On the other hand, our study shows broadly in line with Zachariadis et al. (2001) that it takes roughly ten years or more after the introduction of an emission standard to reach an equal level of average fleet emissions. This delay shows one of the inherent limitations with regard to the effectiveness of road transport policy. Although each new emission standard may introduce significant NO_x reductions over the one it replaces, it takes several years before a substantial portion of the fleet complies with the new emission standard. This leads to a rather gradual reduction in emissions from road transport compared to stationary sources, where new regulations have a more instant effect after their implementation. In order to fully account for the delay in compliance, it is important to note that introduction of new technologies was in some cases accompanied by HDVs and passenger cars emitting much higher in real-world operation than the emission standard level (Hausberger and Rexeis, 2004; Ntziachristos and Samaras, 2000), due to loopholes in the type-approval procedure.

This analysis shows that diesel consumption in vehicles increased substantially between 1990 and 2005. For the purpose of abating CO₂ emissions, this development is very welcomed, but from the air quality perspective, the “dieselization” hampered a more rapid NO_x abatement. If the increase in fuel consumption since 1990 in passenger cars had been met by an increase in gasoline rather than diesel, this would have resulted in around 30% lower NO_x emissions today (2005). The above considerations led us to conclude that the policy aimed at reducing NO_x from the transport sector has not been as effective as the ambition level.

Some issues for future considerations are identified. Due to the increase in diesel consumption, primary NO₂ emissions may be increasing in Europe, despite the over-

10728

all reduction in NO_x . In diesel exhausts, excess oxygen may lead to much higher NO_2/NO_x ratios (50%) in vehicles equipped with oxidation aftertreatment (diesel oxidation catalyst or catalyzed filter) for PM control (AQEG, 2006) than for gasoline three-way catalytic converter cars (less than 5%). Current evidence shows that ambient concentrations of NO_2 do not decrease at the same rate as NO_x in various European hotspots (Lambrecht, 2007; Carslaw et al. 2007), mainly due to the increasing NO_2 ratio in late diesel technology vehicles. Hourly NO_2 concentration limit values become mandatory in Europe starting from 2010 (EC Daughter Directive 99/30/EC). The proportion of primary NO_2 in vehicle exhausts may need to be addressed in future NO_x inventories. Another effect of the increase in diesel consumption in road transport is that less non-methane volatile organic pollutants (NMVOC) emissions are emitted from this sector. The average NO_x/VOC emission ratio for PC and LDV has increased by a factor 2 between 1990 and 2005 according data officially reported to the UNECE. The impact on tropospheric ozone production of the above EU wide changes in emission ratios from road transport should be further assessed by air quality modelling.

It has been demonstrated here that implied emissions factors for NO_x in 2005 are sometimes even higher than the emission standard requested 15 years before. Although the slow vehicle replacement rate is responsible for a large part of this deviation, an equally significant part originates from the discrepancy of real-world operation emissions and emission standards. The next stage of emission standards expected in Europe by 2009–2010 (Euro 5) will further reduce NO_x emissions from both gasoline and diesel vehicles. This will be achieved with both in-cylinder measures and aftertreatment devices, such as selective catalytic reduction (SCR) systems and lean- NO_x catalysts. It needs to be made sure that these devices will be effective over the complete or, at least, a large portion of the engine operation range, to avoid excessive off-cycle emissions. Emission control regulations in the future should therefore more effectively address off-cycle emissions, e.g. by introducing a type-approval test covering a wider range of engine operation modes.

This paper does not analyse the implications for NO_x emissions from the transport

10729

sector by introducing larger proportion of biofuels in accordance with the EC biofuel directive (Directive 2003/30/EC), nor the contribution from international shipping on European NO_x emission levels, but these are nevertheless important subjects for future studies.

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10730

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10731

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10732

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10733

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10734

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10735

Table 1. Emission factors for nitrogen oxides related to fuels and sectors.

| Fuel | Activity | Emission Factor (g NO ₂ /kg) |
|---------------------------------|--|--|
| Hard coal (25 PJ/Tg) | Thermoelectric power plants | 9 |
| | Gas works | 1 |
| | Coke production ¹ | 1.5 |
| | Industry sector | 7 |
| | Transport (railways) | 2 |
| | Other (residential) | 2 |
| Brown coal (11 PJ/Tg) | Thermoelectric power plants ² | 2–8 |
| | Industry sector ² | 1.5–5 |
| | Other (residential) | 2 |
| Residual fuel oil (40 PJ/Tg) | Thermoelectric power plants | 10 |
| | Industry sector | 8 |
| | Refineries | 6 |
| | Transport | 6 |
| | Other (residential) | 8 |
| Gas/diesel oil (43 PJ/Tg) | Thermoelectric power plants | 6 |
| | Industry sector | 6 |
| | Transport (heavy duty vehicles) ³ | 30–50 |
| | Agriculture (machinery) ³ | 40–50 |
| | Residential | 2 |
| | Other | 5 |
| Jet fuel (46 PJ/Tg) | Aviation | 10 |
| Kerosene (46 PJ/Tg) | Residential | 1 |
| Gasoline (46 PJ/Tg) | Transport (passenger cars) ³ | 20–30 |
| LPG (46 PJ/Tg) | Other (residential) | 4 |
| Natural gas (48 PJ/Tg) | Thermoelectric power plants | 0.6 |
| | Industry sector | 0.4 |
| | Other (residential) | 0.3 |
| Wood (19 PJ/Tg) | Residential fire places | 1.9 |

¹ Including gas produced and burnt in association with the coke production (see text)

² Depending on fuel quality and combustion technology in the respective countries

³ Depending on combustion concept and operation conditions

10736

Table 2. Nitrogen oxides trends per European country 1980–2005 (Unit: Gg NO₂). Percentage contribution from road transport in brackets. Countries highlighted in – Grey: Officially reported data. Bold italics: Reported data completed by independent estimates. Stars: RAINS data, interpolation and extrapolation. Normal: EDGAR data, interpolation and extrapolation.

| | 1980 | 1985 | 1990 | 1995 | 2000 | 2005 |
|----------------------------|------------------|------------------|------------------|------------------|------------------|------------------|
| Albania* | 25 (55) | 29 (57) | 23 (37) | 16 (65) | 22 (62) | 25 (65) |
| Armenia* | 15 (44) | 45 (49) | 60 (41) | 18 (20) | 31 (59) | 38 (61) |
| Austria | 249 (45) | 236(46) | 211(47) | 192(49) | 204(54) | 225(58) |
| Azerbaijan | 85 (45) | 93 (41) | 93 (21) | 85 (8) | 76 (4) | 85 (4) |
| Belarus | 234 (43) | 238 (42) | 285 (33) | 232 (28) | 208 (41) | 184 (30) |
| Belgium | 442 (46) | 325 (56) | 382 (48) | 372(47) | 330 (46) | 293(43) |
| Bosnia and Herzegovina* | 66 (48) | 73 (38) | 73 (28) | 51 (23) | 53 (28) | 52 (34) |
| Bulgaria | 357 (50) | 375 (49) | 363(38) | 264(33) | 184(31) | 233(39) |
| Croatia | 60(48) | 73 (38) | 86(38) | 60(45) | 72(43) | 69(40) |
| Cyprus | 13(56) | 14(58) | 16(42) | 19(44) | 23(43) | 17(39) |
| Czech Republic | 937 (21) | 831 (22) | 742 (19) | 413 (43) | 398 (42) | 278 (35) |
| Denmark | 273(26) | 291(32) | 274(38) | 264(37) | 207(39) | 186(37) |
| Estonia | 67 (43) | 74 (41) | 74(41) | 38(42) | 35(38) | 32(34) |
| Finland | 295(36) | 275(44) | 299(53) | 258(51) | 235(45) | 77(32) |
| France | 1942(43) | 1726(51) | 1840(59) | 1654(60) | 1405(52) | 1207(45) |
| Georgia | 121 (43) | 140 (41) | 64 (57) | 13 (11) | 30 (10) | 32 (12) |
| Germany | 3334(35) | 3276(38) | 2861(47) | 2170(53) | 1817(55) | 1443(45) |
| Greece | 242 (40) | 306(39) | 299(36) | 320(39) | 328(37) | 317(34) |
| Hungary | 273(41) | 263(42) | 276(42) | 193(45) | 194(52) | 203(62) |
| Iceland | 21 (21) | 21 (20) | 26 (21) | 27 (21) | 28 (22) | 29 (27) |
| Ireland | 73(36) | 91(40) | 121(36) | 123(38) | 130(40) | 116(37) |
| Italy | 1606(40) | 1661(41) | 1943(46) | 1808(51) | 1373(51) | 1173(46) |
| Kazakhstan | 164 (21) | 179 (19) | 179 (18) | 162 (8) | 119 (8) | 151 (8) |
| Latvia | 61 (43) | 67 (41) | 67(30) | 40(37) | 38(42) | 41(43) |
| Lithuania | 152(36) | 166(34) | 158(34) | 65(36) | 49(51) | 58(58) |
| Luxembourg* | 23 (40) | 21 (40) | 20 (44) | 32 (75) | 33 (80) | 29 (80) |
| Malta | 12 (39) | 15 (38) | 14 (20) | 13 (22) | 12 (27) | 12 (24) |
| Netherlands | 583(40) | 589(44) | 558(47) | 468(45) | 384(45) | 344(42) |
| Norway | 181(32) | 213(31) | 213(35) | 212(30) | 212(21) | 197(18) |
| Poland | 1229 (38) | 1500 (26) | 1581 (25) | 1121 (28) | 838 (27) | 811 (28) |
| Portugal | 166(33) | 166(37) | 246(32) | 278(32) | 287(39) | 281(36) |
| Republic of Moldova | 58 (43) | 66 (42) | 131 (26) | 79 (26) | 27 (30) | 31 (28) |
| Romania* | 523 (27) | 542 (24) | 527 (23) | 400 (22) | 331 (25) | 346 (34) |
| Russian Federation | 3280 (37) | 3600 (33) | 3600 (31) | 2563 (36) | 2357 (40) | 2795 (43) |
| Serbia and Montenegro* | 118 (48) | 145 (38) | 165 (32) | 133 (30) | 137 (36) | 149 (36) |
| Slovakia | 226 (28) | 201 (29) | 215 (21) | 174 (23) | 109 (31) | 97 (38) |
| Slovenia | 51 (52) | 53 (50) | 63 (58) | 66 (65) | 60 (61) | 58 (59) |
| Spain | 1045 (33) | 954 (37) | 1178 (41) | 1254 (39) | 1349(39) | 1405 (34) |
| Sweden | 404 (44) | 426 (41) | 314 (55) | 280 (54) | 231 (49) | 205 (41) |
| Switzerland | 170(61) | 179(71) | 158(59) | 122(53) | 104(53) | 86(49) |
| TFYR of Macedonia* | 37 (48) | 47 (38) | 46 (23) | 35 (30) | 39 (34) | 30 (33) |
| Turkey* | 364 (43) | 483 (39) | 691 (42) | 789 (44) | 942 (36) | 932 (42) |
| Ukraine* | 1598 (15) | 1754 (13) | 1753 (12) | 1245 (15) | 861 (22) | 960 (26) |
| United Kingdom | 2772 (36) | 2728 (40) | 2966 (45) | 2384 (46) | 1897 (43) | 1627 (34) |
| Total | 23 944 (36) | 24 550 (36) | 25 256 (38) | 20 507 (41) | 17 809 (42) | 17 059 (39) |

10737

Table 3. Comparison between EMEP and EURONOX 1980 and 1985 national total and road transport emission data (Unit: Gg NO₂)¹.

| Country/Inventory | 1980 | | | | 1985 | | | |
|-----------------------|----------------|---------|----------------|---------|----------------|---------|----------------|---------|
| | National total | | Road transport | | National total | | Road transport | |
| | EMEP | EURONOX | EMEP | EURONOX | EMEP | EURONOX | EMEP | EURONOX |
| Albania | 25 | 25 | 14 | 14 | 29 | 29 | 16 | 16 |
| Austria | 249 | 231 | 112 | 130 | 229 | 236 | 109 | 148 |
| Belgium | 442 | 371 | 202 | 144 | 325 | 325 | 182 | 157 |
| Bulgaria | 357 | 357 | 177 | 177 | 375 | 375 | 182 | 182 |
| Cyprus | 13 | – | 7 | – | 14 | – | 8 | – |
| Denmark | 273 | 290 | 70 | 95 | 291 | 291 | 92 | 115 |
| Finland | 295 | 228 | 105 | 96 | 275 | 222 | 120 | 119 |
| Former Czechoslovakia | 1163 | 616 | 264 | 168 | 1033 | 597 | 239 | 165 |
| Former USSR | 5835 | 5720 | 1807 | 2483 | 6421 | 6143 | 1818 | 2529 |
| Former Yugoslavia | 332 | 371 | 162 | 177 | 391 | 464 | 155 | 175 |
| France | 1942 | 1931 | 827 | 986 | 1726 | 1793 | 879 | 1113 |
| Germany | 3334 | 3390 | 1163 | 1381 | 3276 | 3509 | 1231 | 1595 |
| Greece | 242 | 242 | 98 | 98 | 306 | 307 | 120 | 128 |
| Hungary | 273 | 297 | 111 | 174 | 263 | 287 | 111 | 175 |
| Iceland | 21 | 12 | 4 | 10 | 21 | 15 | 4 | 13 |
| Ireland | 73 | 89 | 26 | 49 | 91 | 77 | 36 | 55 |
| Italy | 1606 | 1429 | 646 | 763 | 1661 | 1576 | 678 | 958 |
| Luxembourg | 23 | 21 | 9 | 14 | 21 | 24 | 8 | 19 |
| Malta | 12 | – | 5 | – | 15 | – | 6 | – |
| Netherlands | 583 | 508 | 234 | 234 | 589 | 494 | 262 | 252 |
| Norway | 181 | 143 | 58 | 107 | 213 | 151 | 66 | 107 |
| Poland | 1229 | 1147 | 466 | 294 | 1500 | 1192 | 385 | 274 |
| Portugal | 166 | 135 | 55 | 77 | 166 | 149 | 56 | 88 |
| Romania | 523 | 568 | 141 | 203 | 542 | 565 | 130 | 172 |
| Spain | 1045 | 866 | 341 | 405 | 954 | 995 | 351 | 515 |
| Sweden | 404 | 286 | 177 | 169 | 426 | 278 | 176 | 190 |
| Switzerland | 170 | 138 | 104 | 104 | 179 | 170 | 127 | 127 |
| Turkey | 364 | 356 | 157 | 164 | 483 | 513 | 189 | 255 |
| United Kingdom | 2772 | 2266 | 989 | 884 | 2728 | 2261 | 1099 | 1018 |
| Total | 23 944 | 22 033 | 8530 | 9598 | 24 550 | 23 030 | 8834 | 10 658 |

¹ Former USSR includes emissions from Armenia, Azerbaijan, Belarus, Estonia, Georgia, Kazakhstan, Latvia, Lithuania, Republic of Moldova, Russian Federation and Ukraine. Former Czechoslovakia includes Czech Republic and Slovakia. Former Yugoslavia includes Bosnia and Herzegovina, Croatia, Serbia and Montenegro, Slovenia and The former Yugoslav Republic (TFYR) of Macedonia.

10738

Table 4. Emission standards for road transport in Europe post–1992.

| Emission Standard | Regulation | Impl. Year ⁽¹⁾ | NO _x (g/km) or (g/kWh) | NO _x (Gg/PJ) (Converted) | Main technology improvements over preceding step |
|------------------------------|---------------------|---------------------------|-----------------------------------|-------------------------------------|--|
| Gasoline PCs and LDVs (g/km) | | | | | |
| Euro 1 | 91/441/EC | 1992 | 0.62 ⁽²⁾ | 0.25 | Closed-loop TWC ⁽³⁾ |
| Euro 2 | 94/12/EC | 1996 | 0.35 ⁽²⁾ | 0.14 | Faster light-off |
| Euro 3 | 98/69/EC | 2000 | 0.15 | 0.06 | Faster light-off and twin lambda control |
| Euro 4 | 98/69/EC | 2005 | 0.08 | 0.03 | Faster light-off and improved lambda control |
| Euro 5 & 6 | EC 715/2007 | 2010–2015 | 0.06 | 0.02 | Improved aftertreatment materials, deNO _x for direct injection vehicles |
| Diesel PCs and LDVs (g/km) | | | | | |
| Euro 1 | 91/441/EC | 1992 | 0.90 ⁽²⁾ | 0.44 | Improved combustion |
| Euro 2 | 94/12/EC | 1996 | 0.67 ⁽²⁾ | 0.32 | Oxidation catalyst |
| Euro 3 | 98/69/EC | 2000 | 0.50 | 0.24 | Two oxidation catalysts, high pressure injection |
| Euro 4 | 98/69/EC | 2005 | 0.25 | 0.12 | Precise injection and pressure control |
| Euro 5 | EC 715/2007 | 2010 | 0.18 | 0.09 | Diesel particle filters |
| Euro 6 | EC 715/2007 | 2010 | 0.08 | 0.04 | deNO _x , presumably SCR ⁽³⁾ |
| HDVs (g/kWh) | | | | | |
| Euro I | 91/542/EEC | 1992 | 8.0 | 0.84 | Improved combustion |
| Euro II | 91/542/EEC | 1996 | 7.0 | 0.74 | Electronic engine control |
| Euro III | 1999/96/EC | 2000 | 5.0 | 0.56 | High pressure injection |
| Euro IV | 1999/96/EC | 2005 | 3.5 | 0.40 | EGR, precise injection control |
| Euro V | 1999/96/EC | 2008 | 2.0 | 0.25 | Cooled EGR ⁽³⁾ or SCR |
| Euro VI | Only draft proposal | 2014 | 0.4 | 0.05 | Presumably SCR+DPF ⁽³⁾ |

¹ For LDVs and HDVs. For LDVs, the implementation date is roughly one year later than PCs to allow for calibration of new technology.

² Regulations set a standard for the sum of HC and NO_x emissions. The value quoted in the table is an inferred value based on typical HC/NO_x split for the particular vehicle technology.

³ TWC: Three-way catalytic converter; SCR: Selective catalytic reduction; EGR: Exhaust gas recirculation; DPF: Diesel particle filter

10739

Table 5. Implied emission factor for 2005 (Unit: Gg NO₂/PJ).

| | PC | LDV | HDV |
|----------------|------|------|------|
| Austria | 0.20 | 0.22 | 0.79 |
| Denmark | 0.25 | 0.32 | 0.81 |
| Estonia | 0.30 | 0.32 | 0.47 |
| France | 0.27 | 0.35 | 0.39 |
| Germany | 0.14 | 0.29 | 0.76 |
| Lithuania | 1.04 | 0.45 | 0.60 |
| Macedonia | 0.75 | 0.37 | 0.98 |
| Netherlands | 0.19 | 0.31 | 0.72 |
| Norway | 0.18 | 0.14 | 0.55 |
| Poland | 0.43 | 0.44 | 0.81 |
| Portugal | 0.29 | 0.34 | 0.61 |
| Romania | 0.76 | 0.53 | 0.65 |
| Slovakia | 0.31 | 0.34 | 0.60 |
| Slovenia | 0.31 | | |
| Spain | 0.39 | 0.39 | 0.43 |
| Switzerland | 0.11 | 0.30 | 0.76 |
| United Kingdom | 0.22 | 0.25 | 0.62 |

10740

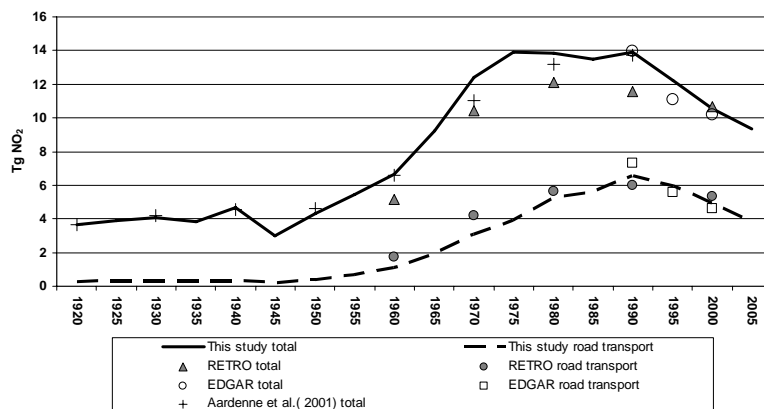


Fig. 1. Comparison between this study and the van Aardenne et al. (2001), RETRO and EDGAR inventories for OECD Europe as defined in EDGAR.

10741

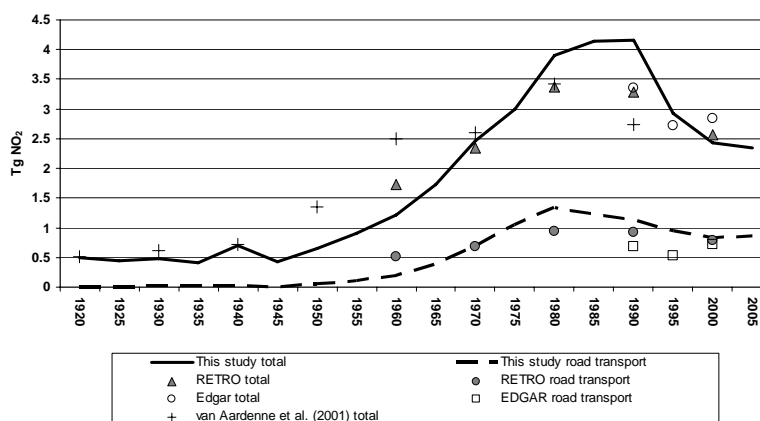


Fig. 2. Comparison between this study and the van Aardenne et al. (2001), RETRO and EDGAR inventories for East Europe as defined in EDGAR.

10742

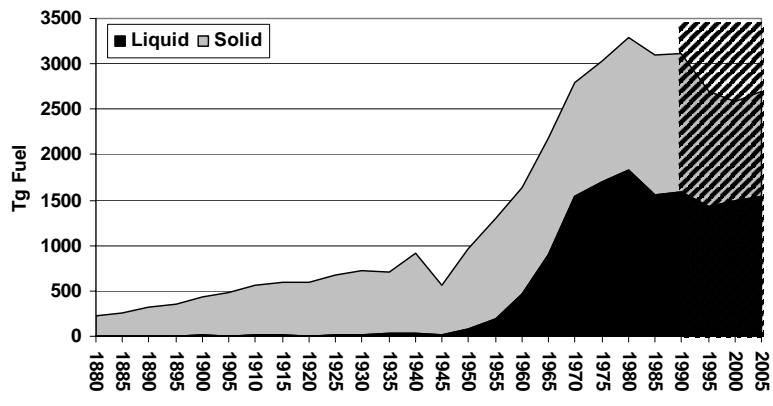


Fig. 3. European solid and liquid fossil fuel consumption 1880–2005. Data from the GAINS model 1990–2005 (Tg fuel/year).

10743

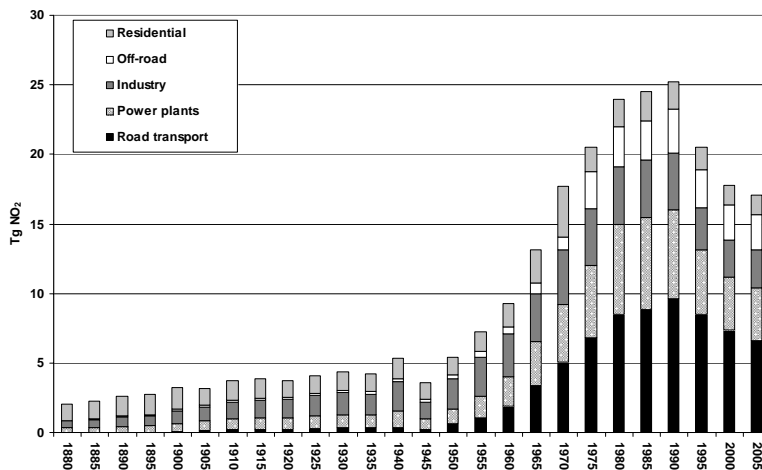


Fig. 4. Sector trends in European NO₂ emissions 1880–2005 (Unit TgNO₂).

10744

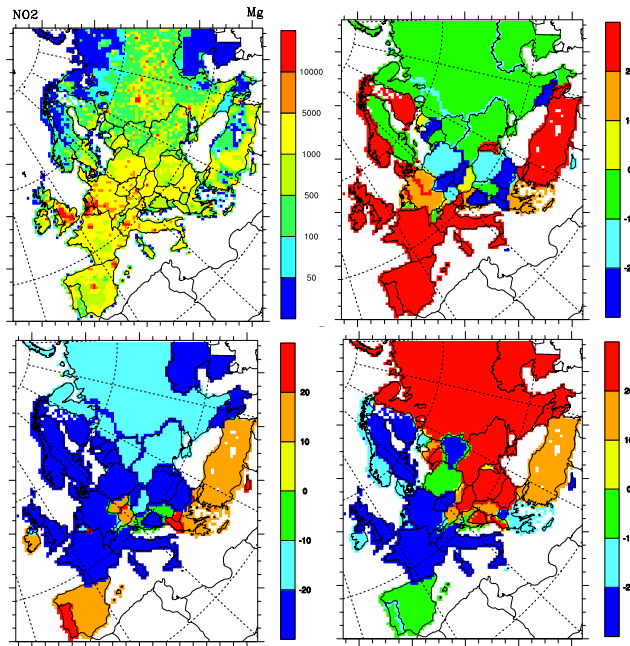


Fig. 5. Road transport emissions of NO₂ in 2005 (top left). Unit Mg. Difference in road transport emissions between 1980 and 1990 (top right), 1990 and 2000 (bottom left), 2000 and 2005 (bottom right). A negative number indicates a reduction. Unit: Percent.

10745

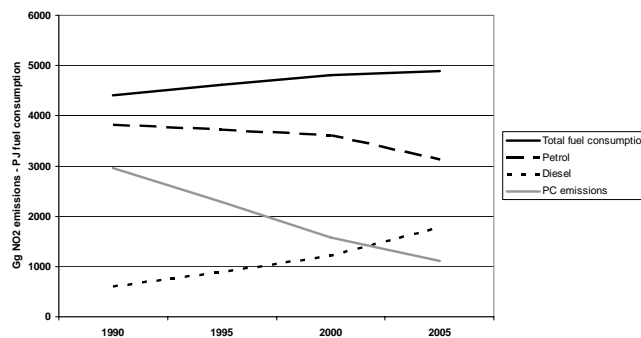


Fig. 6. Trends in Western European fuel consumption and emissions from Passenger Cars (Austria, Denmark, France, Germany, Netherlands, Norway, Portugal, Spain, Switzerland and United Kingdom).

10746

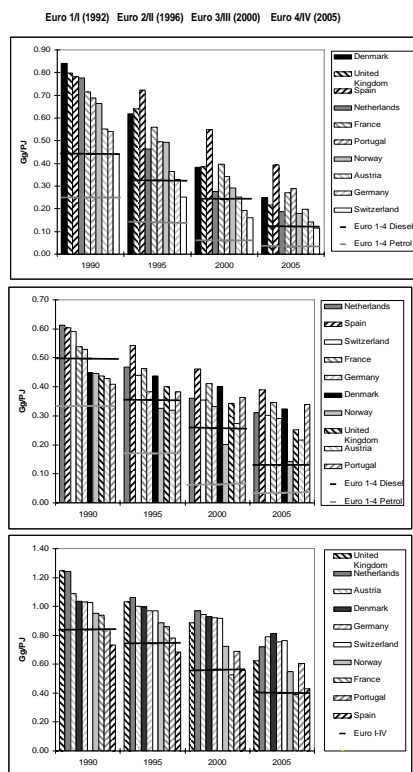


Fig. 7. Implied emission factors 1990–2005 for Passenger Cars (top), Light Duty Vehicles (middle) and Heavy Duty Vehicles (bottom) compared to the Euro standards.

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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 SO₂ 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 trans-boundary, 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 (SO₂), oxides of nitrogen (NO_x), ammonia (NH₃) 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 SO₂ 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 SO₂, both in the gas phase (by OH) and in the aqueous phase (by O₃, H₂O₂,

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HO₂NO₂ and metals). Lifetime is of the order of 1–2.5 d for SO₂ and 4–6 d for sulphate (Koch et al., 1999; Chin et al., 2000a; Rasch et al., 2000; Rotstajn 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 Holtlag 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 O₃–NO_x–VOC cycle. In addition, five sulphur components (DMS, SO₂, sulphate, H₂S 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 SO₂ 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). H₂S, volcanic SO₂ and biomass burning of SO₂ are all taken from Spiro et al. (1992). All these

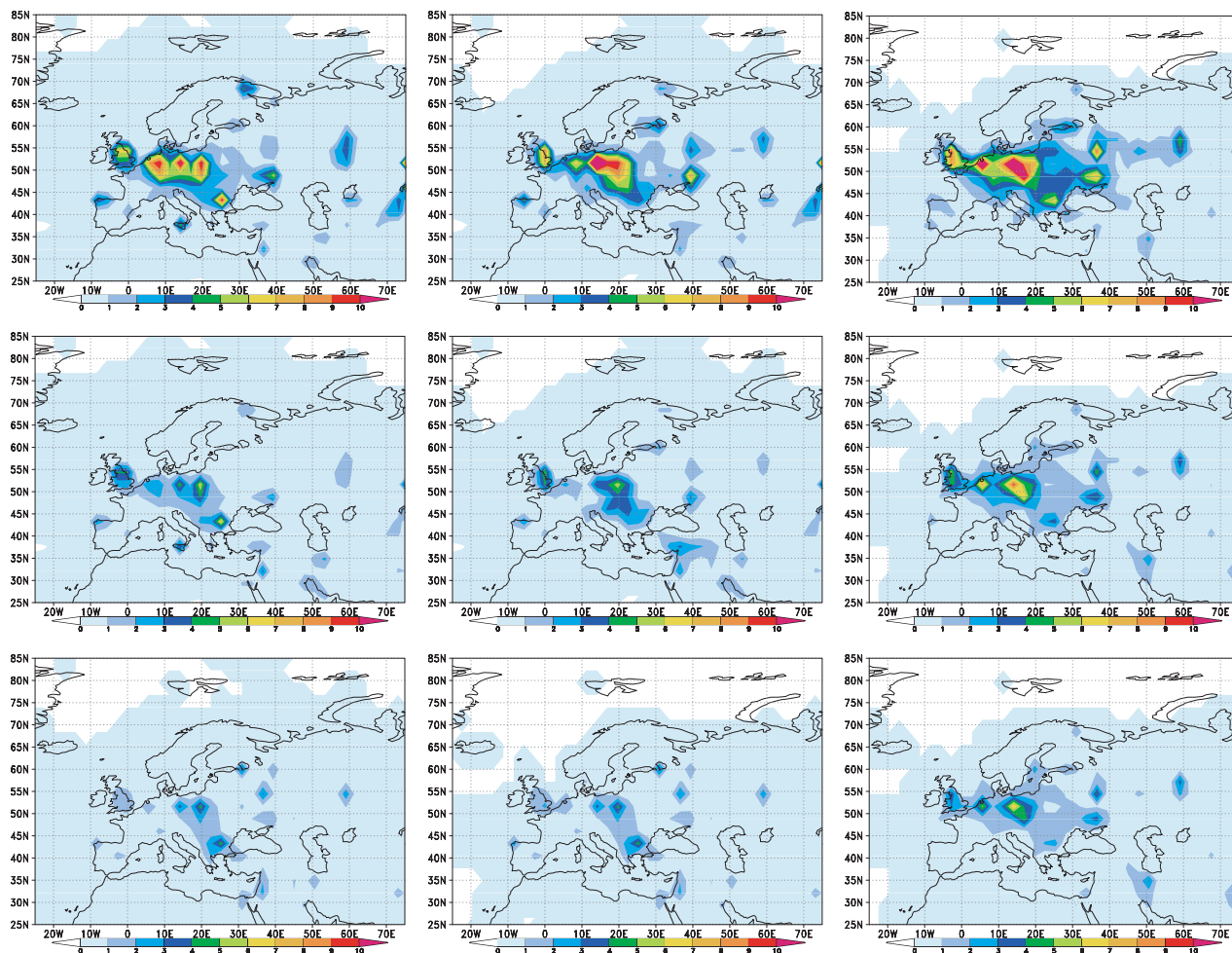


Fig. 1. SO₂ emissions, annual mean, 1985 (upper row), 1995 (middle), and 2000 (lower row) using EMEP (left-hand column), GEIA/EDGAR/AEROCOM (middle) and Smith et al. (right-hand column) emission inventories. Unit: 10^{12} molec $m^{-2}s^{-1}$.

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 ($5.6^\circ \times 5.6^\circ$, 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 SO₂ 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 |

^aEMEP provide emissions for Europe only.

^bNo emissions of oxidants provided for 2000, use EDGAR 1995.

^cGEIA 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 SO₂ 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 |

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,

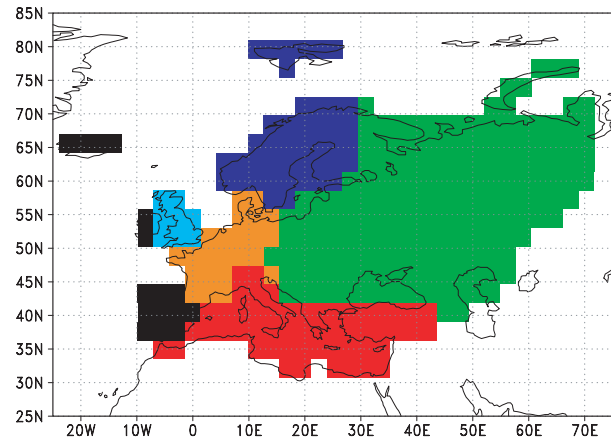


Fig. 2. Regions used in this study. See Table 3 for colour codes.

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 |

^aThere 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 SO₂ and sulphate (see Table 3

for colour codes). The model both under and overestimates the SO₂ observations for 1985 (i.e. there is a wide spread in the plotted points) while it strongly overestimates SO₂ observations for 1995 and 2000. The EMEP runs show higher correlation coefficients ($r = 0.49$ – 0.63) for SO₂ 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 SO₂). 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 SO₂/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

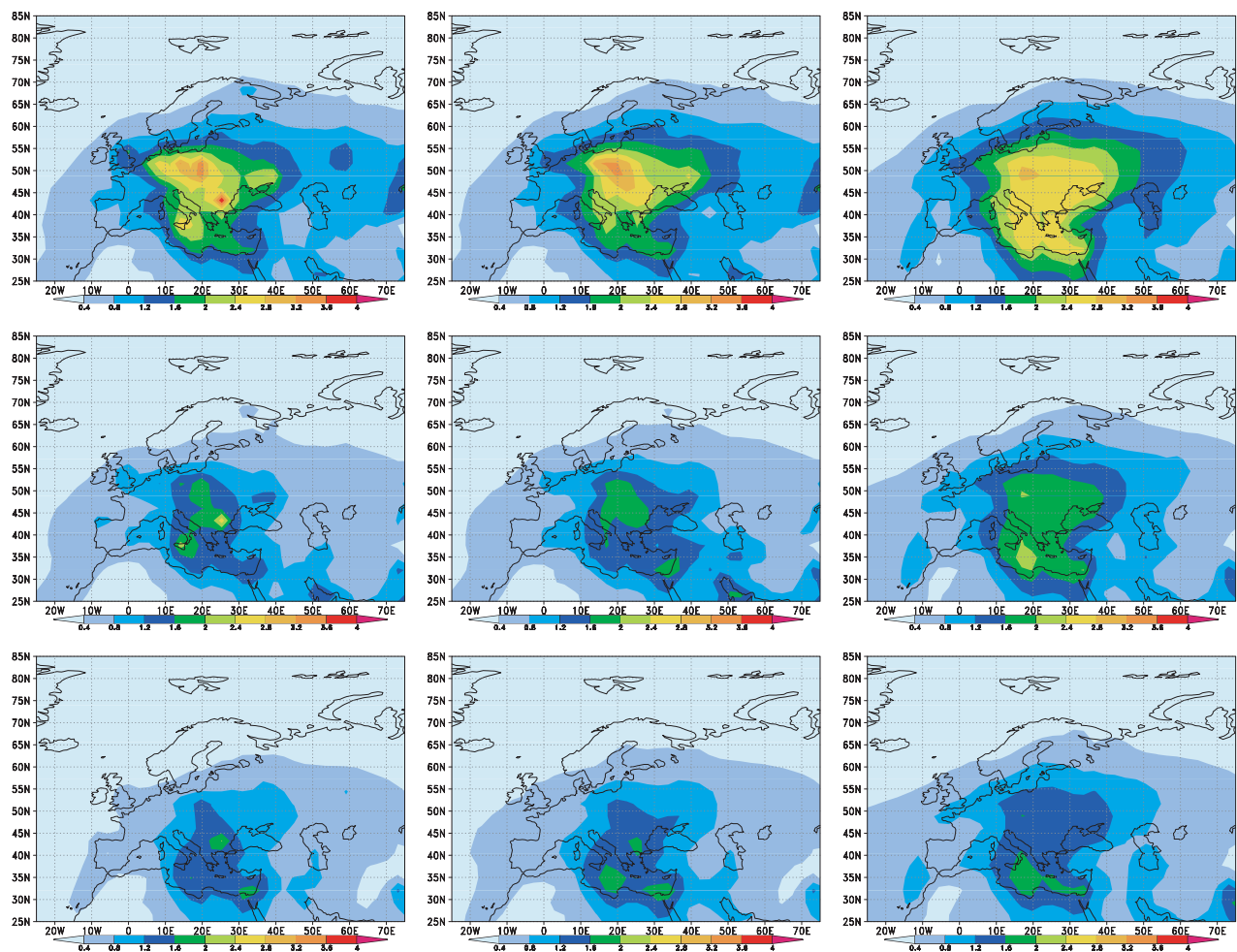


Fig. 3. SO₄²⁻ model concentration, annual mean, 1985 (upper row), 1995 (middle), and 2000 (lower row) using EMEP (left-hand column), GEIA/EDGAR/AEROCOM (middle) and Smith et al. (right-hand column) emission inventories. Unit: $\mu\text{g m}^{-3}$.

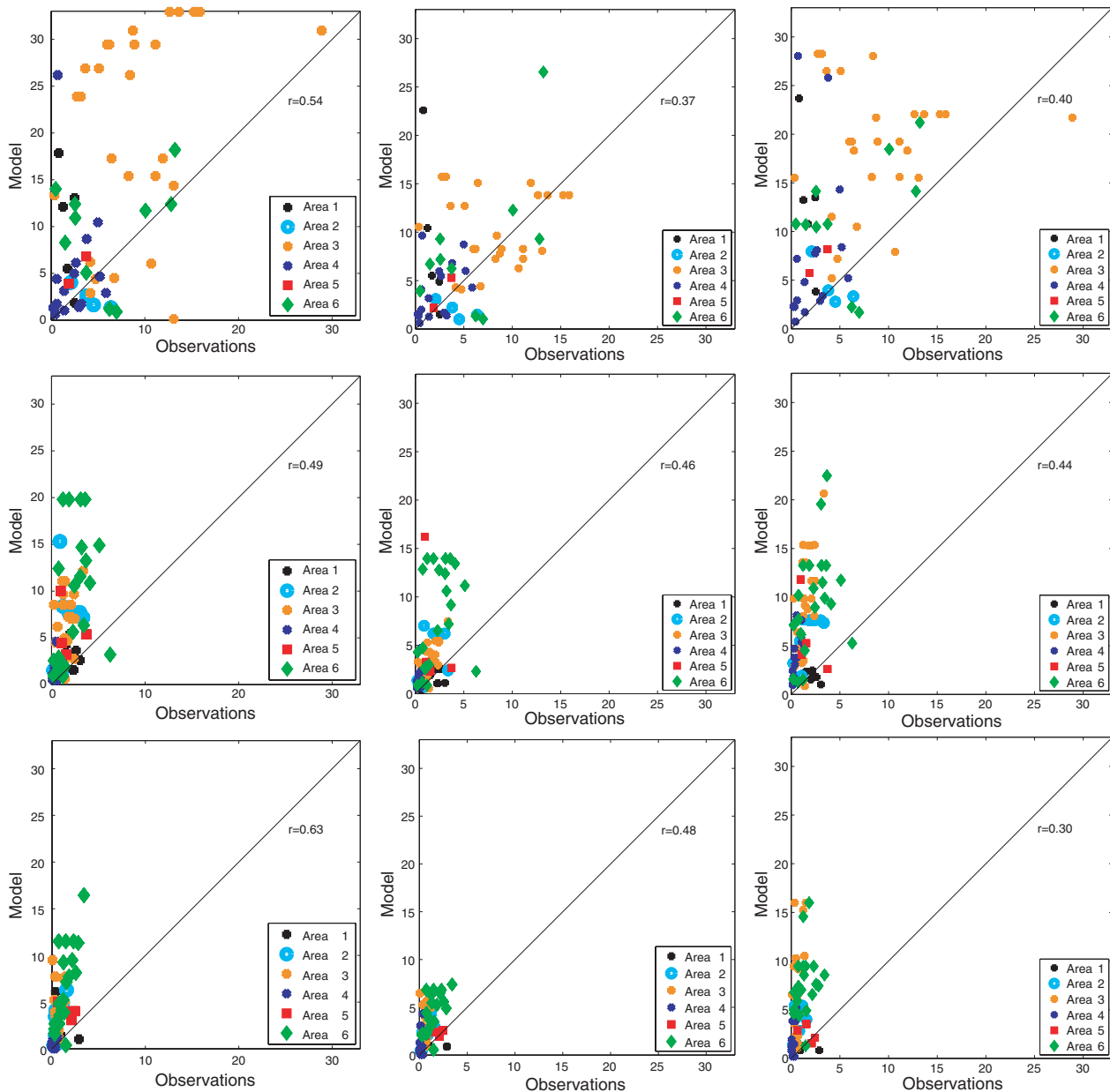


Fig. 4. SO_2 model concentration versus observations, annual mean, 1985 (upper row), 1995 (middle) and 2000 (lower row) using EMEP (left-hand column), GEIA/EDGAR/AEROCOM (middle) and Smith et al. (right-hand column) emission inventories. Unit: $\mu\text{g m}^{-3}$. Annual mean of the observations is 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 oxida-

tion 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 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

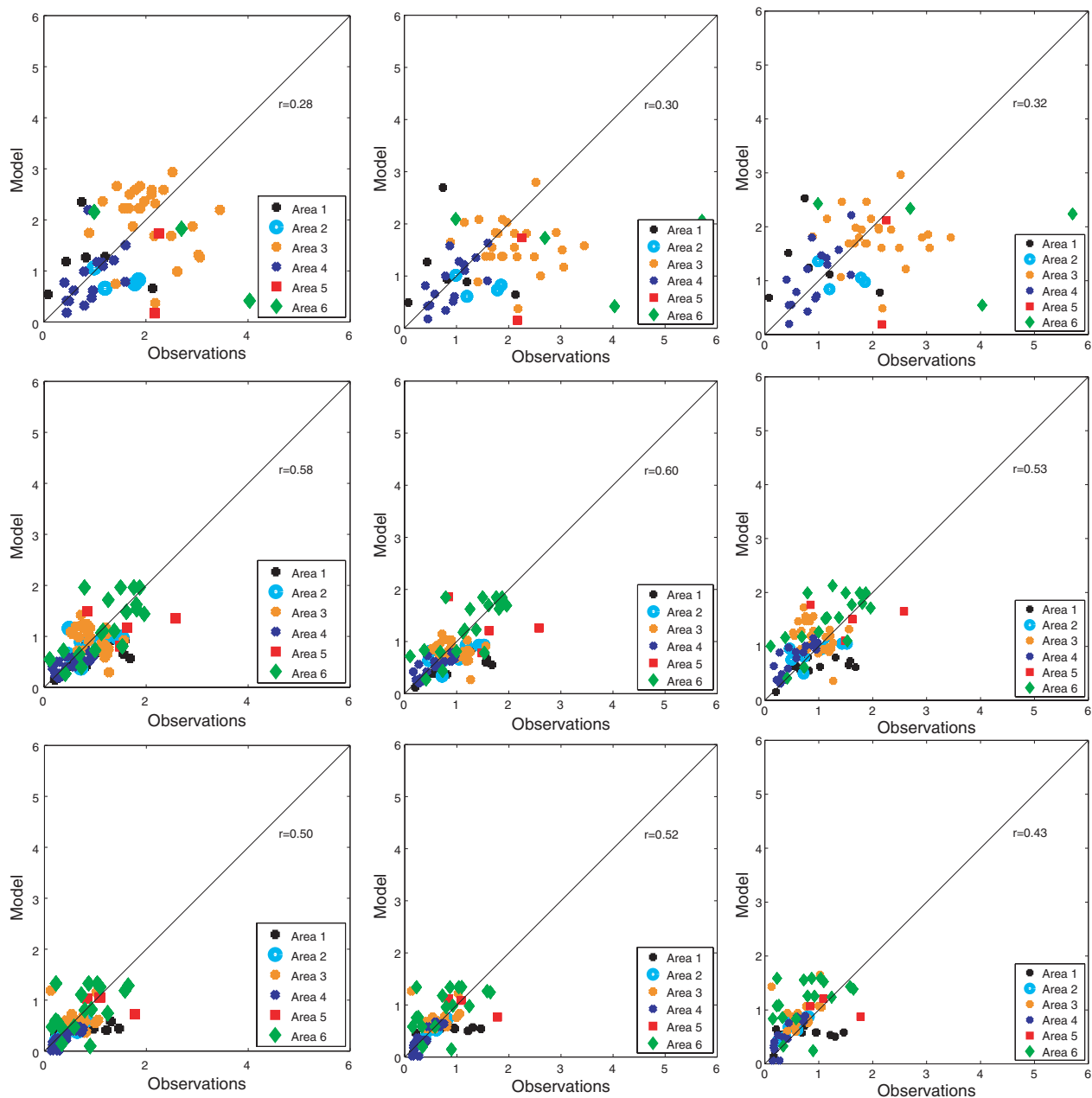


Fig. 5. Same figure, but for SO_4^{2-} . Unit: $\mu\text{g m}^{-3}$. Maximum value in plot: 5.9.

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

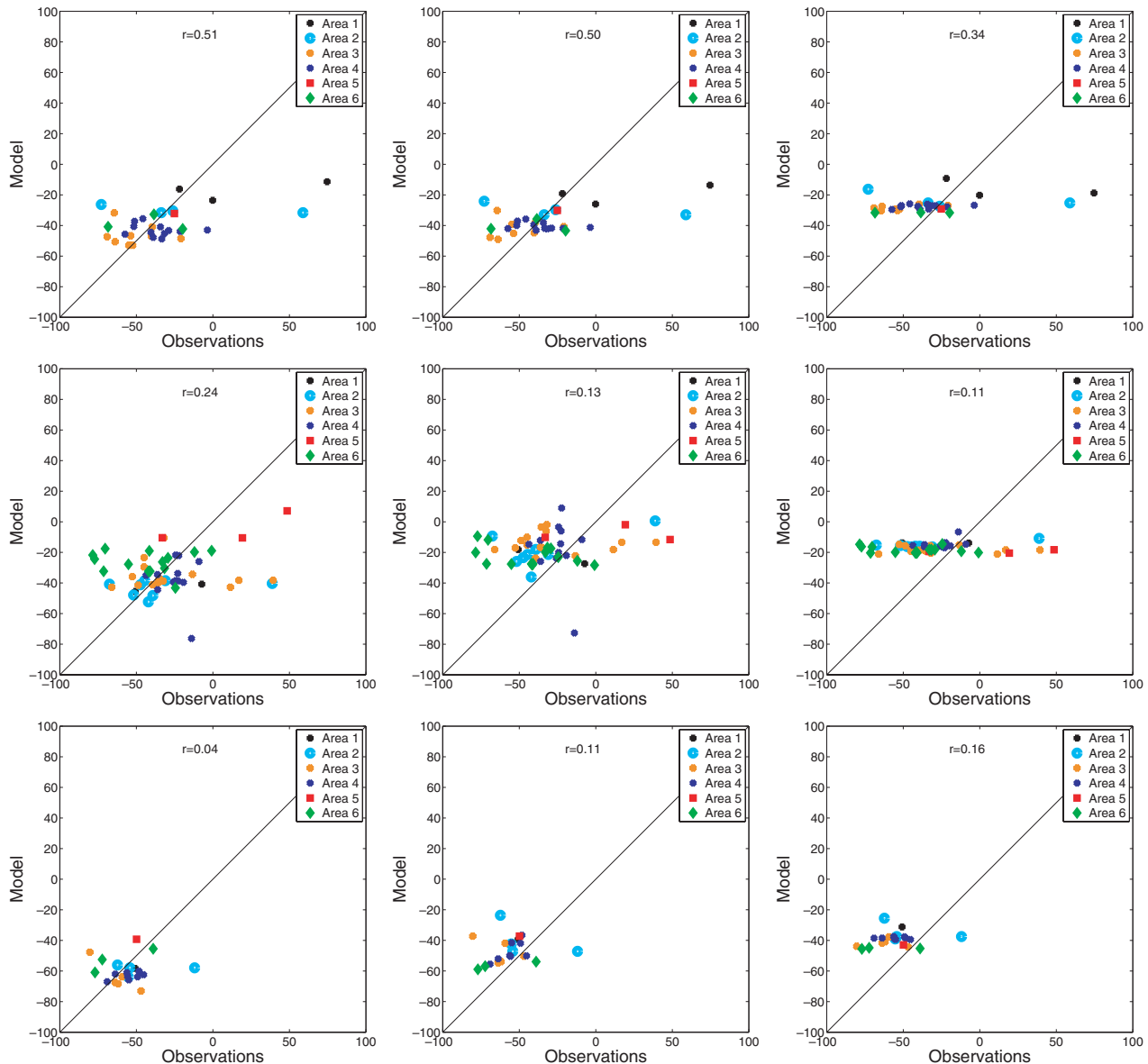


Fig. 6. Trends of sulphate, percent change, 1985–1995 (upper row), 1995–2000 (middle) and 1985–2000 (lower) using EMEP (left-hand column), GEIA/EDGAR/AEROCOM (middle column) and Smith et al. (right-hand column) emission inventories, model results versus observations. Correlation coefficients are included.

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 SO₂, 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 SO₂ 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. SO₂⁴⁻/Ox. SO₂ 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 SO₂ 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 SO₂ 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. SO₂ 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 SO₂. This is a pattern seen in many previous studies of the sulphur cycle. An oxidation limitation leading to high SO₂/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

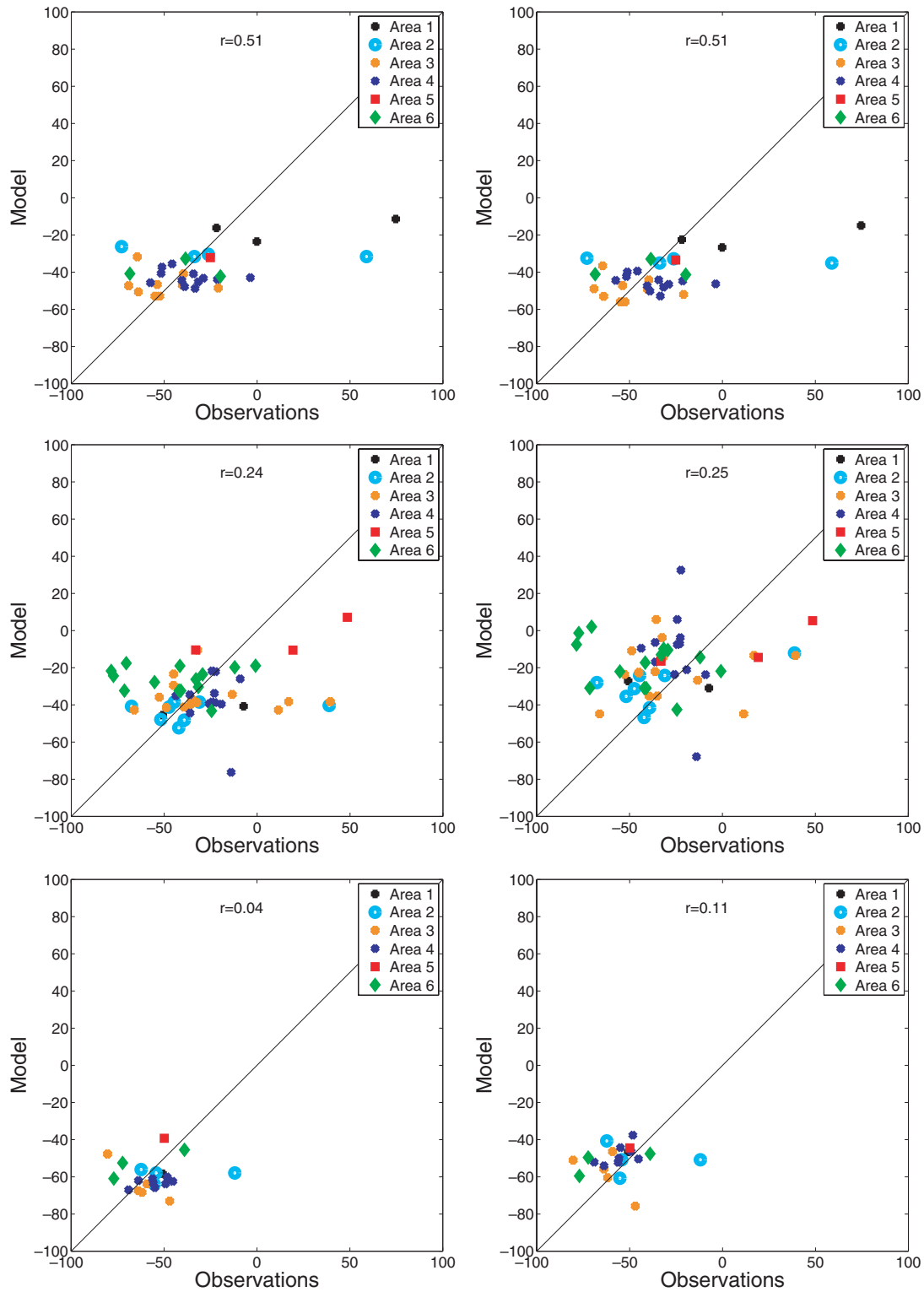


Fig. 7. Trends of sulphate, percent change, 1985–1995 (upper row), 1995–2000 (middle) and 1985–2000 (lower) using EMEP emission inventories, 2000 meteorology (left-hand column), 1997 meteorology (right-hand column), model results versus observations. Correlation coefficients are included.

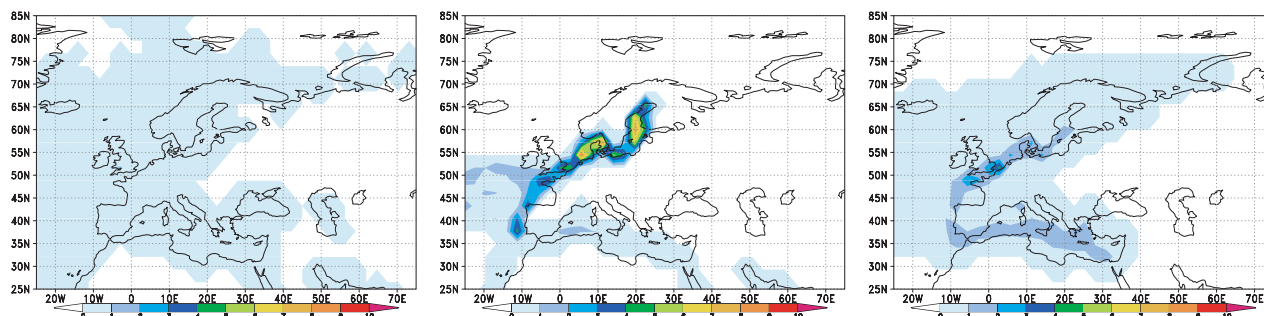


Fig. 8. Ship emissions for the year 2000, AMVER (left), AEROCOM (middle) and EMEP (right) inventories. Unit: 10^{10} molec. m^{-2} s^{-1} .

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 coastal 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 socio-economic 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.

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Modeling historical long-term trends of sulfate, ammonium, and elemental carbon over Europe: A comparison with ice core records in the Alps

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[1] The regional EMEP chemical transport model has been run for the 1920–2003 period and the simulations compared to the long-term seasonally resolved trends of major inorganic aerosols (sulfate and ammonium) derived from ice cores extracted at Col du Dôme (CDD, 4250 m above sea level, French Alps). Source-receptor calculations have been performed in order to allocate the sources of air pollution arriving over the Alps. Spain, Italy, France, and Germany are found to be the main contributors at CDD in summer, accounting for 50% of sulfate and 75% of ammonium. In winter more European wide and trans-Atlantic contributions are found. The relative impact of these sources remains similar over the whole Alpine massif although transport from US and emissions from Spain contribute less as we move eastward from CDD, toward other alpine ice core drill sites like Colle Gnifetti (CG) in the Swiss Alps. For sulfate, the CDD ice core records and the simulated trends match very well. For ammonium, the trend simulated by the model and the summer ice core record are in reasonable agreement, both showing greater changes in ammonium concentrations than would be suggested by historical ammonia emissions. Motivated by a such good agreement between simulations of past atmospheric concentrations and ice core records for inorganic aerosol species, we also use the model to simulate trends in elemental carbon for which less information on past emission inventories are available.

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1. Introduction

[2] Human activities have greatly increased the inputs of sulfur, nitrogen and carbonaceous compounds to terrestrial and aquatic ecosystems since preindustrial times. It is well established that sulfur and nitrogen deposition affects ecosystems, directly or indirectly via chemical changes induced in soils, ground waters and surface waters. The effects tend to result from deposition accumulated over decades, thus it is important to know the long-term loads. In addition, inorganic aerosols (especially sulfate) and carbonaceous aerosols (especially black or elemental carbon) are very important components of the Earth's radiation balance [Intergovernmental Panel on Climate Change, 2001]. Today, several historical emission inventories exist for SO₂, NO_x, NH₃, CO, VOCs and carbonaceous aerosols

[e.g., Mylona, 1996; Lefohn et al., 1999; van Aardenne et al., 2001; Asman et al., 1988; Ito and Penner, 2005]. These inventories can be used as inputs to chemical transport models, thereby estimating historical concentrations and depositions of anthropogenic species. However, there are several uncertainties involved in this approach and the outcome of models needs to be validated against observations. Historic records of nitrogen and sulfur compounds in the European atmosphere are scarce and restricted to the most recent decades. SO₂ and sulfate background concentrations have been monitored in Europe at several sites since around 1980, for instance through the EMEP Programme (Cooperative Programme for Monitoring and Evaluation of the Long-Range Transmission of Air pollutants in Europe). At a few European sites, total nitrate (nitrate aerosol plus nitric acid) and NH_x (ammonia plus ammonium aerosols) have been sampled since 1990 but, even today, the spatial locations of such sites are mainly restricted to areas in central-west and north Europe. Only a few continuous short-term deposition data sets are available [Oden, 1976; Brimblecombe and Stedham, 1982], but from 1955 to 1979, sulfate, ammonium and nitrate wet deposition data were collected within the European air chemistry network

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[Söderlund and Granat, 1982; Rodhe et al., 1984; Söderlund et al., 1985].

[3] In addition to these atmospheric data covering the recent decades, numerous long-term trends of sulfate, nitrate and ammonium have been extracted from Alpine ice cores. The first advantage of such records is that they extend back to the preindustrial era. Second, the vertical distribution of pollutants in the atmosphere is an important constraint for chemical transport models and atmospheric data gained at such high-elevation continental sites are useful for analysis of model dynamics. Alpine ice cores are well suited for the purpose of reconstructing past atmospheric concentrations as they are surrounded by highly populated and industrial European areas. Unfortunately, several studies have shown that for such small areas the snow accumulation characteristics of glaciers, and their changes upstream to the drill sites, can affect the quality of the ice records in view of reconstructing past atmospheric changes [Preunkert et al., 2000]. For instance a key point is the separation of winter and summer trends that are representative of different air masses (tropospheric background in winter versus boundary layer in summer). One of the aims within the CARBOSOL project [Legrand and Puxbaum, 2007] is to evaluate the extent to which ice core records obtained at a high-elevated Alpine site can help to constrain poorly known past emission inventories of carbonaceous aerosols. In this paper we examine ice core trends of major inorganic aerosols at the Alpine site Col du Dôme (CDD, 4250 m above sea level, French Alps) in the light of model simulations. We simulate summer and winter atmospheric concentrations of sulfate and ammonium over the 1920–2003 time period by using the regional EMEP model and past emissions inventories of SO₂ and NH₃. Here summer is defined as April–September in the modeling and winter as October to March. The summer/winter criteria for observations are discussed in section 2.1. The ammonium in the ice cores originates both from gas phase ammonia and particle ammonium (NH_x), and the sum of the species have been extracted from the model calculations in the comparison of model results and inverted ice core concentrations. However, the fraction of ammonia is very low (model calculations give around 5%), thus it is basically a comparison of ammonium aerosol concentrations.

[4] The model calculations also consider past changes of NO_x, CO and VOCs. Simulated concentrations are first compared to present-day atmospheric observations made in recent years at the Vallot Observatory (VO), located nearby the CDD ice core drill site. Thereafter, simulated trends of atmospheric concentrations are compared to winter and summer ice core records which were inverted into historical atmospheric concentrations by using local firn to air (FAR) relationships established by Preunkert et al. [2001] for present-day summer and winter conditions. Further, the influence of the variability in meteorological conditions on atmospheric concentrations is investigated. By establishing so-called source-receptor relationships from model calculations, source apportionment is achieved at CDD as well as at another alpine drill sites located further east, CG, where ice records are also available but far less seasonally resolved than at CDD. Finally, an attempt is made to investigate the accuracy of past emission inventories of elemental carbon in the light of the long-term trend

of this carbonaceous aerosol component extracted by Legrand et al. [2007] in the CDD ice cores.

2. Observations

2.1. Ice Core Records

[5] The main characteristics of the CDD ice core records we use here have been presented by Preunkert et al. [2000]. The dating of the CDD ice core was established by counting annual layers along the ammonium profile and using various time horizons. Time horizons are gained from ¹³⁷Cs measurements which permit identification of the 1986 (Tchernobyl event) and those of 1954 and 1963 (atmospheric nuclear tests) layers as well as the calcium record of Saharan dust horizons (1997, 1947, and 1936/1937). The annual layer counting was found to be in good agreement with the three ¹³⁷Cs horizons suggesting a precise dating (± 1 year) over the 1954–1994 time period. On the basis of Saharan dusts events, it was shown that the uncertainty in snow deposits from 1925 to 1954 is ± 5 years. Each annual snow layer was divided in two parts corresponding to winter and summer snow accumulation. The dissection cutting has been based on the ammonium profile [Preunkert et al., 2000]. The frequency distribution of ammonium concentrations in the upper part of the CDD ice core (covering the 1981–1994 time period) indicates a bimodal distribution with a low concentration mode below 10 ppb and a second mode centered on 200–300 ppb. The boundaries of the winter half year snow pack have been identified by requiring at least 3 consecutive samples to significantly exceed the 10 ppb level.

[6] At CG, no seasonal dissection was attempted but CG snow deposits are usually made by summer layers, because of the preferential loss of winter snow by wind erosion at this site [Wagenbach et al., 1988].

2.2. Present-Day Measurements

[7] In order to gain reliable year-round data on the chemical aerosol composition above 4000 m elevation, an automatic aerosol sampler was developed and deployed since 1999 at VO (4360 m a.s.l., French Alps) located close to Col du Dôme [Preunkert et al., 2002]. The flow rate of the device is 3 L STP per minute and each aerosol sample covers 7–10 days in summer and 20 days in winter.

[8] These atmospheric data were also useful to investigate FAR relationship needed to invert ice core data in terms of atmospheric concentrations (see section 8).

3. Emissions

3.1. Anthropogenic Inorganic Emissions

[9] Anthropogenic emission data of SO₂, NO_x, NH₃, CO and VOCs from 1980 to 2003 used in the model simulations are based, as far as possible, upon emissions officially reported per emission sector and grid by Parties to the Convention on Long-Range Transboundary Air Pollution [e.g., Vestreng et al., 2004]. For the period prior to 1980, we have used three different sources of information. For CO, NH₃ and VOCs, historic emissions estimated by van Aardenne et al. [2001] were available globally per sector on a $1 \times 1^\circ$ resolution. The EDGAR-HYDE sectors used by van Aardenne et al. [2001] were converted to emissions per

country and to the emission sectors used by EMEP, the so-called SNAP system (Selected Nomenclature for Air Pollution [Vestreng and Klein, 2002]). Scaling factors per country and sector were used to scale the EMEP 1980 emissions backward in time, but ensuring that the country totals are kept. In this way, the better resolution ($\sim 50 \times 50 \text{ km}^2$) of EMEP data could be kept along with the evolution of the historic emissions. In the data from *van Aardenne et al.* [2001], spatial distribution over the years are only different when the relative amount within the sectors change. Thus we lose no information when applying only the scaling factors as the sector information is kept. For SO_2 and NO_x we used the emissions from *Myllona* [1996] and V. Vestreng and A. Semb (Nitrogen oxides emission inventories over Europe since the pre-industrial era, manuscript in preparation, 2007), respectively. Emission scaling factors were defined in the same way as for NH_3 , VOCs and CO. The historical emissions for NO_x and SO_2 were available from 1880 to 1985, for the countries with country borders as they were historically. For instance, emissions are not available separately for the countries within the former Soviet Union. Therefore the countries in the former Soviet Union are scaled with the same factor. East and West Germany are scaled separately back to 1950, but as the sum before. The areas corresponding to Czech Republic and Slovakia are scaled with emissions for former Czechoslovakia. Slovenia, Croatia, Bosnia and Herzegovina, Serbia and Montenegro and the Former Yugoslav Republic of Macedonia are scaled by using the historic emissions of former Yugoslavia. 5% of the SO_2 emissions were assumed to be primary sulfate emissions based on measurements of primary emissions of particulate sulfate in the mid-70s [Shannon et al., 1980]. The different sources of sulfur emissions show large variability in emitted fraction of sulfate, thus the fraction has probably changed somewhat over the years. However, since there is not enough data to give a description of the changes in the primary sulfate emission fraction during the last hundred years, we have kept this estimate for the full period.

[10] Emissions are distributed temporally according to monthly and daily (Sunday to Saturday) factors derived from data provided by the University of Stuttgart (IER). These factors are specific for each pollutant, SNAP sector and country, and thus account for, e.g., the very different climates and hence energy use patterns in different parts of Europe. For instance, SO_2 emissions from combustion in energy and transformation industries (stationary sources), which are the largest sector for SO_2 , are typically a factor 2–3 higher in winter than in summer for northern European countries, and typically more uniform over the year for southern European countries. In contrast, ammonia emissions, which predominantly originate from agriculture activities, peak in early spring, with an additional peak in autumn for countries that have two sowing seasons.

[11] The heights of the stacks have changed significantly during the last century, which we have taken into account by defining a “tall stack” period (from 1955 to present-day) and a “low stack” period (prior to 1955). In the “tall stack” period, the power plant emissions are assumed to have effective emission heights between 180–1100 m, with the peak of the distribution of effective emission heights of about 400 m. In the “low-stack” period, the emissions from

power plants and industry in each model layer are moved one model layer closer to the ground, corresponding to a peak in the effective emission height distribution of about 200 m.

3.2. Elemental Carbon (EC) Emissions

[12] Elemental carbon (EC, or black carbon (BC); the terms are often used interchangeably, and are difficult to define [e.g., *Gelencsér*, 2004]) is an important component of the carbonaceous aerosol. The EC data used in the 2002 simulation is based on a new inventory for annual national emissions developed by *Kupiainen and Klimont* [2007]. Estimates of EC, organic carbon (OC) and PM_{10} emissions were based upon an extensive review of the literature, and further checked for consistency with estimates of fine $\text{PM}_{2.5}$ included previously in the model. This inventory is identical to that used in the CARBOSOL carbonaceous modeling studies [Simpson et al., 2007; Tsyro et al., 2007].

[13] The data assembled for the historical SO_2 and NO_x inventories discussed above, together with additional activity statistics, were combined with appropriate emission factors for EC. The range of emissions factors for EC found in the literature varies considerably, however (factor 10 or more [e.g., *Cooke et al.*, 1999; *Bond et al.*, 2004; *Kupiainen and Klimont*, 2007]), and so the inventories for EC must be acknowledged to have considerably greater uncertainty than those for SO_2 and NH_3 . Our base estimate for 1985 is based upon the *Bond et al.* [2004] emission factors, being the most up to date compilation at the time this study was undertaken, but we additionally consider the variation in emissions factors for the road transport sector. The emission estimates up to 1985 in this study are considered to come from combustion without emission control. The highest emission factors for EC occur for diesel vehicles, followed by hard coal combustion in rail traffic and burning of wood in fireplaces.

[14] Two emission estimates for elemental carbon have been used, one with constant emission factors over time (denoted EC-C), and one where emission factors for road transport are allowed to increase backward in time, from 1985 to 1965, by up to a factor of five, following *Novakov et al.* [2003] (denoted EC-V). Thus the EC-C and EC-V estimates are identical (based upon *Bond et al.* [2004]) for 1985, but they differ substantially around 1970, when the combination of high traffic density and high emission factors leads to maximum emissions.

[15] Open biomass burning (OBB, including forest and agricultural fires) are not implemented in the standard EMEP model used here. *Gelencsér et al.* [2007] performed a source apportionment of $\text{PM}_{2.5}$ organic aerosols over Europe and found median contributions of EC from biomass burning to be about a factor 20–35 lower than the contributions of fossil fuel origin at three different CARBOSOL mountain sites (Puy de Dome, Schauinsland and Sonnblick) in summer. In winter, the contribution was somewhat higher (factor 6–15), but EC from fossil fuel combustion was still predominant.

[16] Using the emission factors suggested by *Andreae and Merlet* [2001] would suggest changes in sulphate, ammonium and EC from OBB of similar orders of magnitude. Thus we conclude that although forest fires may make significant contributions on an episodic basis to concen-

Table 1. European Emissions of EC, 1920–1985, Used in the Calculations^a

| Year | Residential/Domestic Combustion (S2) | Road Traffic (S7) | Other Mobile Sources (S8) | Total |
|------|--------------------------------------|-------------------|---------------------------|-------|
| 1920 | 471 | 0.4 | 83 | 632 |
| 1930 | 466 | 40 | 107 | 707 |
| 1940 | 443 | 72 | 104 | 723 |
| 1950 | 442 | 142 | 113 | 794 |
| 1960 | 427 | 450 | 123 | 1148 |
| 1970 | 328 | 792 | 158 | 1429 |
| 1980 | 257 | 616 | 271 | 1265 |
| 1985 | 253 | 325 | 259 | 949 |

^aEstimate EC-V, with variable emission factors, see section 3.2. Total includes S2, S7, S8 plus other emission sectors. Units Gg. Labels in parentheses refer to emission SNAP sector codes.

trations over Europe, and likely to the Alpine region, the available evidence suggests that European OBB emissions make only small contributions at CDD on the basis of a full summer or winter average.

[17] The annual emissions calculated using method EC-V are shown in Table 1. The European total EC emissions increase from 630 Gg in 1920 to 950 Gg in 1985, and a maximum of 1430 Gg was reached around 1970. On a global scale, the emission inventories from *Ito and Penner* [2005] and *Novakov et al.* [2003] show a gradual increase with emission peaks late in the 1980s and 1990s, in contrast to our estimated emissions which peak around 1970. However, the steepest increase was found outside Europe, namely in China, thus the European and the global trends are hardly comparable. *Novakov et al.* [2003] estimated a factor 4 decrease for Great Britain between 1950 and 1980, which is not far from our estimate (factor 3 decrease).

[18] The moderate trend in our emission estimate masks large changes in especially the transport sectors. Emissions from road transport (denoted S7) increase from less than 0.4 Gg in 1920 to almost 800 Gg in 1970 and 325 Gg in 1985. Emissions from other mobile (denoted S8) sources (including railways), increase from ~ 80 Gg in 1920 to 260 Gg in 1985.

[19] The residential sector (S2) is the far most important of all EC emissions up to approximately 1960, and indeed dominates completely the year 1920 estimate. EC emissions level off from around 1950, followed by a decrease by a factor of 2 from about 1960 as a result of phasing out of coal burning in the residential sector in EU-15 countries and in Scandinavia. In the same period the road (S7) and off-road (S8) emissions increase as a result of increased availability of oil, and a huge increase in the transport of goods and passengers. The emissions of wood burning were found to be relatively constant, at about 60 Gg, throughout the period of the study (because of a fairly stable rural population), hence the relative importance of wood burning emissions are larger for earlier years.

[20] In total, emissions from stationary sources (basically all emissions except emissions in the transport sectors S7 and S8) decrease somewhat from ~ 550 Gg in 1920 to 370 Gg in 1985. The different trends in the transport sectors and emissions from stationary sources are important because of the different seasonality of the emissions: road traffic and other mobile sources emit at rather constant rates over the year, whereas stationary sources have much larger

emissions in wintertime. For instance, emissions from the residential sector (S2) are a factor 5–8 larger in winter than in summer. Thus the change in emissions will impact summer and winter concentrations of EC differently.

[21] Unfortunately, the emission estimate for 2000 and the historical inventory for 1920–1985 were developed by different groups and with somewhat different input data. The year 2000 inventory [*Kupiainen and Klimont, 2007*] has the advantage of using highly detailed statistics on fuel usage and activity data that were simply not available for the historical emissions work. Total European emissions in the historical inventory for 1985 were ~ 950 Gg, whereas emissions for the year 2000 inventory were ~ 680 Gg, i.e., a reduction of $\sim 30\%$.

3.3. Natural Emissions

[22] Biogenic emissions of dimethyl sulfide (DMS) are implemented in the model as monthly averaged emission data derived from *Tarrasón et al.* [1995]. These DMS emissions are treated as SO_2 as inputs to the calculations. Sulfur emissions from volcanoes are included for Italy and treated as point sources at a height determined by the altitude of the volcanoes. Emissions of NO_x from lightning are included as monthly averages on a T21 ($5.65 \times 5.65^\circ$) resolution [*Köhler et al., 1995*]. Biogenic emissions of isoprene and monoterpenes are calculated as a function of land use, temperature and solar radiation, using procedures detailed in *Simpson et al.* [1999, 2003a].

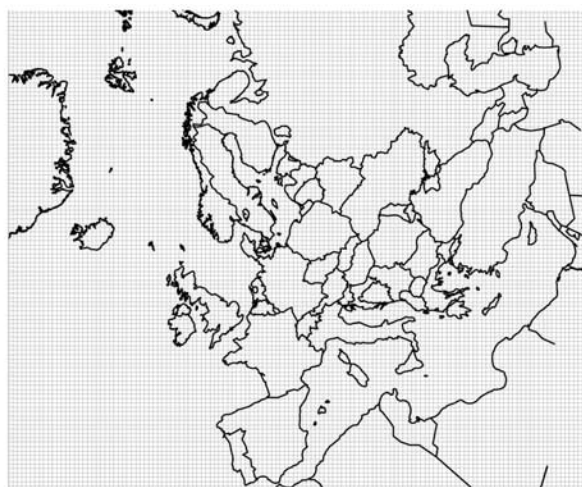
3.4. Ship Emissions

[23] Total releases of SO_2 , NO_x , NMVOCs and CO from ship traffic in the Atlantic Ocean, North Sea, Baltic Sea, Black Sea and Mediterranean Sea are used following *Lloyd's Register of Shipping* [1995, 1998, 1999]. These estimates are of the same magnitude as those derived by ENTEC (Environmental and Engineering Consultancy) for 2000 [*Whall et al., 2002*], thus we assume that they are valid for 2000. In the model calculations we have assumed a 2.5% increase per year, consistent with the increase of international shipping emissions from 1995 to 2000 in the work by *Endresen et al.* [2003]. The ship emissions of SO_2 , NO_x , NMVOCs and CO in the period from 1920 to 1975 have been assumed to be directly proportional to the register tons related to the steam and motor ships as detailed by *Mitchell* [1981].

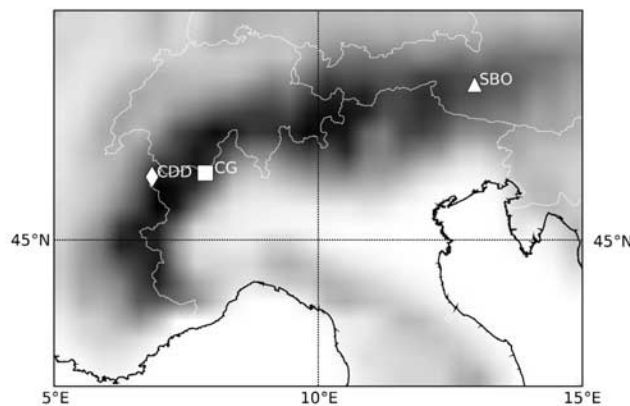
4. EMEP Unified Model Calculations

4.1. Model Description

[24] The Eulerian EMEP model is a multilayer atmospheric dispersion model designed to simulate the long-range transport of air pollution over several years. The model is fully documented by *Simpson et al.* [2003a] and *Fagerli et al.* [2004] and some applications of the model are given by *Fagerli et al.* [2003], *Simpson et al.* [2006a, 2006b] and *Fagerli and Aas* [2007] for sulfur and nitrogen species and by *Jonson et al.* [2006] for ozone and NO_2 . The model domain is centered over Europe but also includes most of the North Atlantic and the north polar regions (Figure 1). The model has 20 vertical layers in σ coordinates below 100 hPa. It is primarily intended for use with a horizontal resolution of $\sim 50 \text{ km} \times 50 \text{ km}$ (at 60°N) in the



(a) EMEP model domain



(b) Location of CDD, CG and SBO

Figure 1. EMEP model domain used for this study and the location of CDD, CG and Sonnblick (SBO).

EMEP polar stereographic grid. The model uses meteorological data from PARLAM (PARAllell version of HIRLAM) [Benedictow, 2002], a dedicated version of the operational HIRLAM model (High Resolution Limited Area Model) maintained and verified at MET.NO. The chemical scheme includes about 140 reactions between 70 species. SO_2 is oxidized to sulfate both in gas phase with OH and in aqueous phase through H_2O_2 , O_3 and O_2 catalyzed by metal ions, assuming a fixed pH value of 4.5. The sulfur chemistry is coupled to the photochemistry, thus changes in the oxidation capacity may change the SO_2 oxidation rate and vice versa. The partitioning between NH_3 and ammonium aerosols is calculated by using the EQSAM module detailed by Metzger *et al.* [2002a, 2002b]. The dry deposition module makes use of a stomatal conductance algorithm originally developed for calculation of ozone fluxes, here applied in the model to all pollutants for which stomatal control is important [Emberson *et al.*, 2000a, 2000b, 2000c; Simpson *et al.*, 2001, 2003b; Tuovinen *et al.*, 2001, 2004]. Parametrization of the wet deposition processes in the

EMEP model includes both in-cloud and subcloud scavenging of gases and particles, using scavenging coefficients.

[25] As a part of the CARBOSOL project, the EMEP chemical transport model has been extended to cover carbonaceous aerosol, including EC [Simpson *et al.*, 2007; Tsyro *et al.*, 2007]. Emissions, and wet and dry deposition are simulated in a similar way as sulfate, although with somewhat lower removal rates to reflect the more hydrophobic nature of EC. Using current emission levels, Simpson *et al.* [2007] and Tsyro *et al.* [2007] found fairly good agreement with current-day levels of EC at sites from the EMEP and CARBOSOL networks.

4.2. Boundary Conditions

[26] Boundary conditions (BIC) for a number of species are described with simple functions. These have been designed to enable concentration values that correspond to observations. The concentrations are adjusted in the vertical and for latitude and time of the year (monthly) to match the observed distributions. The annual cycle of each species is represented with a cosine curve, using the annual mean near-surface concentration, the amplitude of the cycle, and the day of the year at which the maximum value occurs. The parameters used to set the prescribed boundary conditions are described by Simpson *et al.* [2003a] and Fagerli *et al.* [2004].

[27] Our default BIC set is based on measurements from the period around 1980. To account for changes in the concentrations at the boundaries, we have applied two sets of scaling factors. The first set is used to scale the BIC for the years 1980–2003. The scaling factors have been defined on the basis of the EPA (U.S. Environmental Protection Agency) emissions (U.S. Environmental Protection Agency [2000] and updates on their web page, <http://www.epa.gov>) for SO_2 and NO_x emissions for the 1980–2003 period. Although U.S. NH_3 emissions have been relatively stable during this period, SO_2 emissions have decreased by more than 40% and NO_x emissions by approximately 20% (1980–2002, <http://www.epa.gov>), hence the trend in ammonium aerosol will be determined by sulfate and nitrate availability rather than by changes in NH_3 emissions [Fagerli and Aas, 2007]. Therefore the changes in BIC for ammonium aerosols were set by weighting the trend of SO_2 emissions with 2/3 and NO_x emissions with 1/3.

[28] The second set of scaling factors was developed for the period prior to 1980. In winter, CDD is located above the boundary layer most of the time and the source apportionment at present day, which will be presented in section 7, shows that in winter the dominant part of the sulfate (and nitrate) at CDD originates from sources located outside the western boundary of the EMEP domain. Therefore the changes in concentrations in the winter ice core data are a reasonable indicator of the trend in the boundaries and used as a scaling factor backward in time, relative to the prescribed 1980 levels. This is of course not fully true, as emissions from European emissions sources contribute to the deposition at CDD. Moreover, it limits the validation of our model results to summer values, when BICs are far less important. However, it is difficult to find alternatives. There are no other alpine ice core records with separate summer and winter values, and total year records would be inappropriate as contributions from European sources would be

Table 2. Summary of the Model Calculations

| Set | Simulated Years | Meteorology | Emissions |
|-----------------------------|---|-----------------|---|
| <i>Sulfate and Ammonium</i> | | | |
| 1 | – | 1990, 1995–2003 | 1990 |
| 2 | 1920–1980 (every 10th year), 1985, 1990, 1995, 2000 | 1997 | as appropriate |
| 3 | 1920–1980 (every 10th year), 1985, 1990, 1995, 2000 | 2003 | as appropriate |
| 4 | 1980, 1985, 1990, 1995–2003 | as appropriate | as appropriate |
| <i>Elemental Carbon</i> | | | |
| 5 | 2002 | 2002 | 2000 |
| 6 | 1920–1980 (every 10th year), 1985 | 1997 | EC-V and EC-C, appropriate years |
| 7 | 1920–1980 (every 10th year), 1985 | 2000 | EC-V and EC-C, appropriate years ^a |

^aThree additional sets of calculations were performed using EC-V emissions and omitting emissions from sectors S2, S7 and S8 in separate runs.

large. The anthropogenic sulfate trend recorded in Greenland ice cores corresponds to SO₂ emissions from various regions, including North America and Eurasia, thus they do not either represent a unique North American sulfate signal. Another alternative would be to use historical global model calculations, but this was not available to us at the time when this study was carried out. In addition, the global model results also suffer from uncertainties in emissions, oxidation rates and model formulations themselves. Therefore we consider that using the winter CDD ice core data to set BICs represent the most robust approach.

[29] For ammonium, we used the trends of NH₃ emissions from United States derived from *van Aardenne et al.* [2001].

[30] For elemental carbon, background levels are assumed to be zero for this work. At the site Mace Head in Ireland, which is well suited to sampling air masses crossing the Atlantic from North America [*Cavalli et al.*, 2004; *Kleefeld et al.*, 2002], very low concentrations of EC are observed (typically 50 ng m⁻³). *Gelencsér et al.* [2007] performed measurements of ¹⁴C at Sonnblick in winter, at a sampling time where the station was most likely to be in the free troposphere, and found EC concentrations of 20 ng/m³ STP. Compared to the typical summer values sampled at VO (40–50 ng m⁻³ STP) [*Legrand et al.*, 2007], the background would constitute around 40–50%. Although the conditions at Sonnblick were supposed to reflect the tropospheric background, the site might still be influenced somewhat from ground level sources, and the value can be regarded as an upper limit for the background contribution.

[31] In order to investigate the influence of the background values on the EC trends at CDD, the trend in the EC boundary condition values are also needed. However, the data needed to set a reliable trend in the EC boundary conditions are not available. EC ice core data from Colle Gnifetti exist [*Lavanchy et al.*, 1999], but these do not separate between winter and summer. Historical inventories for U.S. EC are available [e.g., *Ito and Penner*, 2005], but are very uncertain.

[32] We are aware that the assumption of zero EC background contribution does introduce some uncertainty in to the comparison of especially the wintertime trends in EC levels. However, rather than adding additional complexity to the comparison by introducing very uncertain parameters, we focus on the more certain summer values and discuss possible implications for the EC winter trends.

4.3. Choice of Model Layer

[33] There are several challenges when setting up a regional model to simulate atmospheric concentrations at high elevated Alpine sites. In winter, these sites are decoupled from the polluted planetary boundary layer and are representative of free tropospheric conditions. In summer, a more efficient convective upward motion of air masses from the polluted boundary layer enhances the atmospheric levels of pollutants. Consequently, concentrations are much higher in summer than in winter at these elevated sites, in contrast to surface concentrations that are more similar over the seasons.

[34] The EMEP Unified model has a horizontal resolution of 50 km × 50 km and thus a rather rough topography. For instance, the grid cell representing the CDD site has a height of 1700 m above sea level. In the vertical, the model has approximately 10 layers within the boundary layer and 10 above (up to 100 hPa). The relative height of CDD is 2661 m, corresponding to model layer 10. Since the model does not resolve the fine-scale structure of the Mount Blanc massif, it does not take into account for instance local meteorological conditions leading to more vertical mixing than the model predicts at this height. Thus the model layer representing the air mass arrival pattern characteristic for CDD is most likely somewhat closer to the ground than suggested by using a direct calculation of the relative height.

[35] We used the air measurements available at the VO site (section 2.2) located close to the CDD ice core drilling site to decide which model layer is appropriate to be used. This was done by selecting the model layer that gave the best representation of the summer to winter ratio of concentrations, since this should reflect the relative height of the model layer compared to the topography. On this basis, model layer 13 (~3500 m) was adopted for model outputs.

4.4. Model Runs

[36] For sulfate and ammonium four different sets of simulations were performed (Table 2). Set 1 consists of ten runs using the same (1990) emissions but different meteorology (1990, and from 1995 to 2003). This set was designed to examine the impact of the meteorological variability on the ice core records at CDD.

[37] Set 2 uses the 1997 meteorology with emissions and boundary conditions for every tenth year from 1900 to 1980, plus 1985, 1990, 1995, and 2000. Set 3 is identical

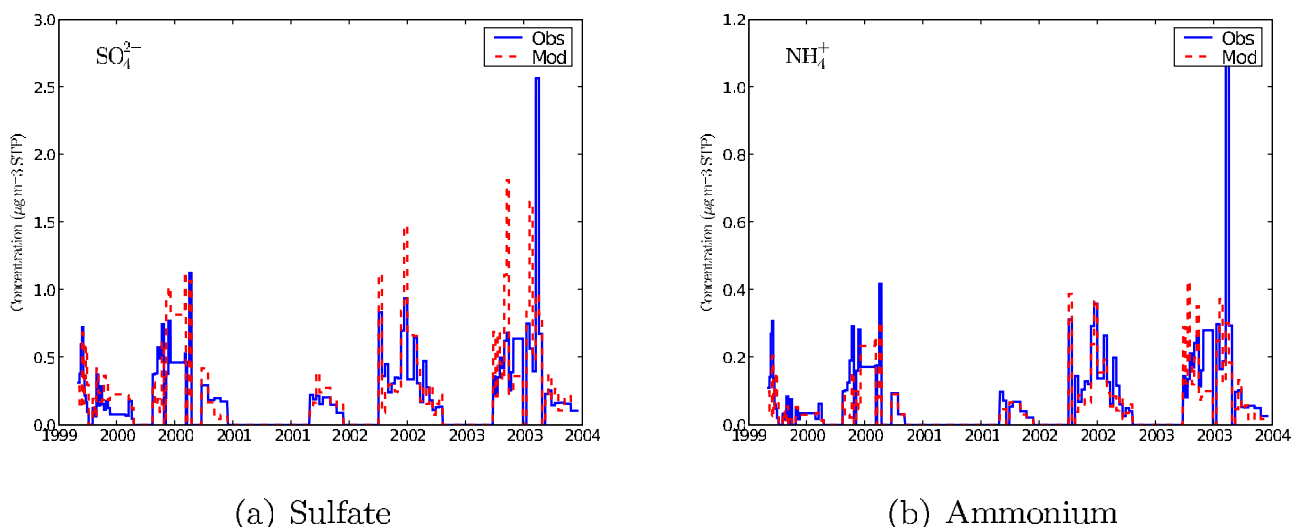


Figure 2. Comparison of simulated and observed concentrations ($\mu\text{g m}^{-3}$ STP) of sulfate and ammonium at Vallot Observatory, 1999–2003.

to set 2 but with 2003 meteorology. These two sets simulate the trend from 1920 to 2000. Because no meteorological data are available back to 1920, we have used two different meteorological years to calculate historical concentrations. The meteorological years 1997 and 2003 were chosen for two reasons. Firstly, after considering the results from set 1, 1997 and 2003 stood out as two extreme years, giving the lowest and highest concentrations at CDD, respectively. Secondly source-receptor matrices were available only for the 2003 meteorology, thus the resulting simulated trend could be related to the sources for this year.

[38] Finally, set 4 uses appropriate meteorology, emissions, and boundary conditions for 1980, 1985, 1990 as well as 1995–2003, and therefore provides the most accurate results. These calculations have also been used in the comparison with available atmospheric data at the VO located close to the CDD drill site.

[39] Three additional sets of simulations were done for EC (Table 2). Elemental carbon at CDD is simulated for 2002 in order to compare with present-day observations of EC at CDD (set 5).

[40] Set 6 simulates EC concentrations for every tenth year from 1920 to 1980 plus 1985 using the 1997 meteorology. Set 7 is identical to set 6 but with 2000 meteorology. The effect of a likely change in emission factors over the past for road transport on the trends of EC is investigated by applying two different emission inventories (EC-C and EC-V, see section 3.2) in both sets. In addition, we estimated the contributions from the different sectors using the 2000 meteorology and the EC-V emissions in sectors S2, S7 and S8 in separate runs.

5. Present-Day Ammonium and Sulfate Levels at CDD

[41] Figure 2 shows the comparison between simulated (model layer 13, ~ 3500 m) and observed concentrations in air for sulfate and ammonium. The model captures very well the observations, both with respect to the seasonality and absolute levels. The lower summer (from April to September)

to winter (from October to March) contrast for sulfate (a factor of less than 3) than ammonium (a factor of 4) seen in observations is also well reproduced by the simulations. This higher summer to winter ratio for ammonium than for sulfate can be explained by the different seasonality of the corresponding surface concentrations. Ammonia emissions result predominantly from agriculture activities, which peak in spring/summer time. Furthermore, the production of ammonium from ammonia is limited by the availability of sulfate and nitric acid (and air temperature), resulting in a rather constant summer to winter ratio of ammonium at the surface. In contrast, the majority of the SO_2 emissions stems from combustion in energy and transformation industries (stationary sources), which are higher in winter than in summer (typically a factor 2–3 in northern Europe, less in southern Europe), resulting in a winter maximum for sulfate surface levels. Therefore the enhanced transport in summer is counteracted by lower surface sulfate levels in that season.

6. Meteorological Variability

[42] Ideally, the calculation of long-term trends should be performed by using the meteorology of the actual years, and for each year up to recent time. In this way, change in weather patterns and interannual meteorological variability would also have been considered in the calculated trends. Unfortunately, there is no such historical archive available in the PARLAM-PS meteorology which we used as input to the EMEP Unified model. Therefore, in the historic calculations of trends we have used the same meteorological conditions for all the years. However, in order to investigate how large the meteorological variability is in the high-elevated Alpine regions, and especially the magnitude of the variability compared to the trends, we have performed a set (set 1, section 4.4) of calculations where we have kept emissions constant (1990 emissions) and varied the meteorology. The resulting sulfate concentrations at CDD, resolved on monthly basis, is shown in Figure 3. On a monthly basis the modeled concentrations vary by as much

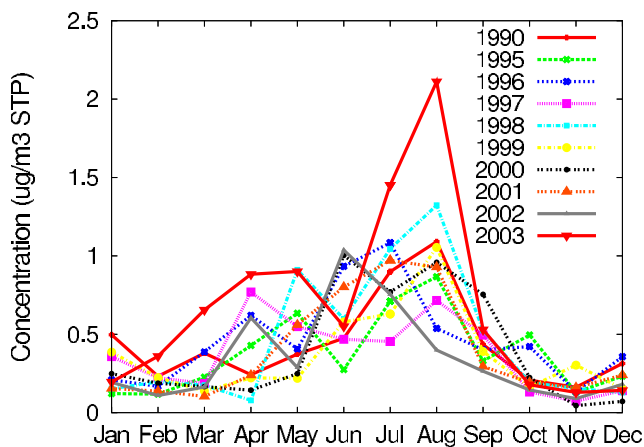


Figure 3. Variability in concentrations ($\mu\text{g m}^{-3}$ STP) of sulfate due to meteorological variability at Col du Dôme. The different lines show the results using different meteorological years. All calculations are done with 1990 emissions.

as a factor of 5 from year to year. For instance the modeled concentrations of sulfate in August, based on 1990 emissions, vary between 0.4 and $2.15 \mu\text{g m}^{-3}$ STP. In January, the concentrations range from 0.15 to $0.50 \mu\text{g m}^{-3}$. The large variability in concentrations caused by the different meteorology is seen for ammonium as well. Of course the impact of the meteorological variability is weaker (a factor of 2–3) when half year summer and winter concentrations, which correspond to the time integrated information extracted from the ice cores, is considered. Nevertheless the magnitude of the meteorological variability still remains significant with respect to long-term changes shown by ice records for the different components (an increase by a factor of 3 for ammonium and 6 for sulfate, see section 8).

[43] *Casty et al.* [2005] analyzed changes in temperature and precipitation in the Alps from 1500 to 2003. Temperature was shown to increase overall from 1900 to the present day, but the most remarkable feature was the much higher temperatures seen for the last 10 years of the reconstruction. The years 1994, 2000, 2002 and 2003 were the warmest years since 1500 in the greater Alpine region.

[44] Trends in precipitation were much less marked. Although some dry and wet periods do occur, the most obvious feature of the precipitation changes were the year to year variability over the last 100 years. Although it seems likely that trends in climate have some influence on the deposition of species at CDD, it is very difficult to assess this contribution. However, the fact that *Casty et al.* [2005] found no significant trends in precipitation amounts, and only moderate changes in temperature, over this period would suggest no dramatic change in air mass origins.

7. Present-Day Source Apportionment at CDD

[45] In this section we investigate the origin of pollutants arriving at CDD by analyzing source-receptor relationships (country to grid) for 2003. We refer to *van Loon et al.* [2005] for a technical description of these kinds of calculations. Numerous discussions on how to deal with source

relationships in Eulerian models, for instance problems related to the nonlinear chemistry, can be found elsewhere [e.g., *Wind et al.*, 2004]. For example, reducing SO_2 emissions in one country may induce higher ammonia deposition there, but lower ammonium deposition in a nearby country since reduced sulfur may limit the formation of ammonium aerosols and thereby the long-range transport of ammonium. The source-receptor matrix for NH_x is thus approximate, but the relationships should give a good indication of the relative contribution of different source areas.

[46] The year 2003 was a rather extreme meteorological year with respect to the summer conditions (very warm and dry summer in Central and South Europe, see *Beniston* [2004] or *Marmier and Langmann* [2007]). It has been shown [*van Loon et al.*, 2005] that the individual contributions from one country to another (or to a grid) can vary substantially for secondary inorganic aerosols from year to year. That is mainly true for the small contributions that in general correspond to inputs related to long-range transport. In contrast, the main contributions are more similar from year to year. For the European Alps, the average meteorological variability of the six largest transboundary contributions to the secondary inorganic aerosols was found to be around 20%.

[47] For sulfate in summer, Spain, Italy, France and Germany are found to be the largest contributors (in total $\sim 50\%$) whereas other significant emission sources (e.g., from Great Britain, GB) are less important (see Figure 4). This supports the attempt made by *Preunkert et al.* [2001] in comparing the long-term trend of sulfate extracted from CDD ice cores with past SO_2 emissions from different countries.

[48] In winter, Spanish, Italian, French and German sources are still the main contributors. However, the individual contribution are smaller, and there are more countries that contribute to the concentrations. For sulfate, the 4 largest contributors (Spain, Italy, France, Germany) accounts for around 50% of the total in summer against 18% in winter. This is again in agreement with the results from *Preunkert et al.* [2001] who found that the recorded increases of sulfate in winter at CDD from 1925 to 1994 matched better with the trend of emissions from total Europe and partly USA. The percentage contribution from the boundaries (mainly hemispheric transport from USA) is much larger in winter compared to summer. This is expected since CDD is a high-elevated site which receives much less pollution from European ground level sources in winter being above the boundary layer most of the time. The absolute contribution of the inorganic aerosols from the boundaries to CDD is approximately equal in summer and winter. This is a result of the boundary conditions for the inorganic aerosols which are essentially independent of altitude and season in the free troposphere, in accordance with observational data [*Warneck*, 2000]. Volcanic emissions also contribute to a substantial part of the observed concentrations in winter. The volcanic emissions are released directly into the free troposphere, thus they are less dependent on the boundary layer mixing, in contrast to anthropogenic pollutants.

[49] For NH_x the source apportionment reveals a similar pattern as for sulfate but the contributions from Spain, Italy,

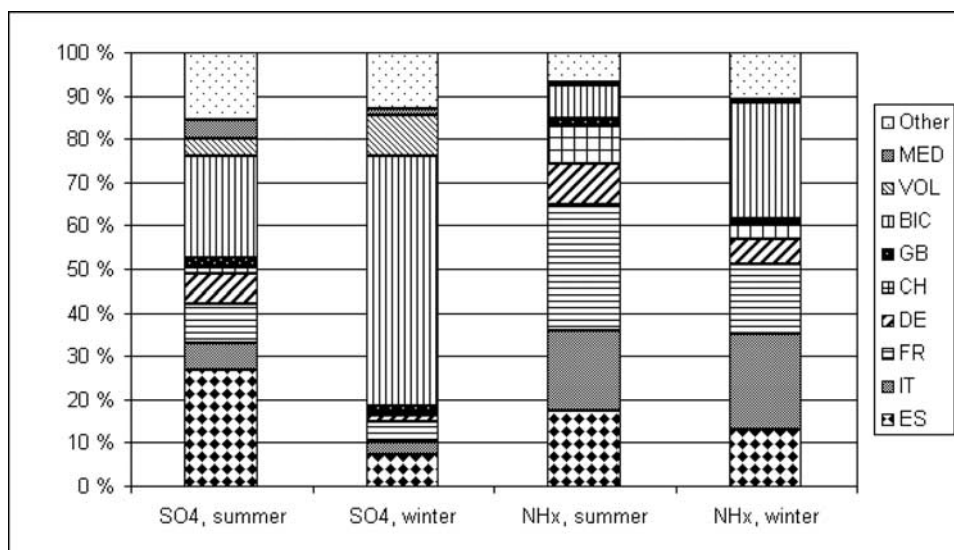


Figure 4. Source allocation for concentrations of sulfate and ammonium plus ammonia (NH_x) for Col du Dôme for 2003: contributions from Spain (ES), Italy (IT), France (FR), Germany (DE), Switzerland (CH), Great Britain (GB), boundary conditions (BIC), international ship traffic in the Mediterranean Sea (MED) and volcanoes (VOL).

France and Germany are higher for NH_x than for sulfate in summer as well as in winter. The reason is that whereas the concentrations of NH_x and sulfate at the boundaries are similar (in volume mixing ratio), the strength of the emission sources of ammonia in these countries are stronger than for SO₂. For instance, France released 750 Gg NH₃ and 505 Gg SO₂ in 2003. Furthermore, there are no ammonia emissions from volcanoes or international ship traffic, which contributes significantly (~10%) to sulfate concentrations at CDD. In total, the sources close to CDD gain a larger weight in the source apportionment for NH_x for 2003.

8. Historical Long-Term Trends of Inorganic Aerosols

[50] In this section we compare simulated past atmospheric concentrations of sulfate and NH_x at CDD with corresponding long-term trends extracted from CDD ice cores.

[51] The seasonal FAR relationships investigated at CDD [Preunkert *et al.*, 2002] were used to invert the ice core concentrations to corresponding ambient air concentrations. These FAR values are, however, relatively uncertain. Furthermore, the dissection of annual snow layers into winter and summer snow layers relies heavily on the ammonium concentration criteria. As described in section 2.1, a criteria of 10 ppb has been used, and the absolute values of the winter ice core concentrations are relatively sensitive to this value, especially for ammonium. Moreover, these winter values represent “low” winter values as high episodes are excluded. The summer values are less sensitive to the ammonium selection criteria, as adding a small part of winter snow (with low concentrations) to the summer snow (with high concentrations) would have a minor influence of the total value. As a consequence, the absolute values of concentrations derived from the ice cores and the model simulated concentrations can be very different, especially

for the winter values. Therefore we do not focus on the agreement of absolute levels but rather on the trends.

[52] Although the winter ice core data are more uncertain than the summer data, we have chosen to include both seasons. The comparison of model simulations and ice core data serves as a consistency check rather than a pure model validation.

8.1. Sulfate Trends

[53] As shown in Figure 5, the simulated trend using level 13 of the model (section 4.3) and the inverted summer ice core record are in excellent agreement with a weak increase between 1920 and 1950, a steep increase toward the maximum around 1980, and then a significant decrease over the last two decades. There is large interannual variability in the ice core values, partly because of meteorological variation (as can be seen from the 1980–2003 model results that has been performed with actual meteorological conditions), and partly because of uncertainty in the extraction of individual ice core values.

[54] As seen in Figures 4 and 6, the peak around 1980 can be explained by the large contributions from Spain, France, Germany and Italy (and the BIC) for which emissions peaked in the 1970s to 1980s, with the later peak being especially prominent for the largest contributor, Spain.

[55] The simulated winter trend in the ice core record at CDD for the period 1980–1994 agree very well with the trend extracted from the ice cores. The winter trend in the ice core record at CDD indicates a regular increase from 1925 to 1980, whilst in the summer record the increase from 1960 to 1980 is four times higher than in the previous period (Figure 5).

[56] In the previous section, we found that BIC (transport from USA), volcanoes and numerous small contributions from countries European wide make up the dominant input of sulfate at CDD in winter in 2003. Because the trend in BIC was set to match the trend in the winter CDD ice core

up to 1980, the similarity of the model simulations and ice core data for the period prior to 1980 is merely a confirmation that the influence of European sources in the earlier period have not been large enough to dominate over the BIC contribution.

8.2. NH_x Trends

[57] As shown in Figure 7, the CDD ice core record show increasing ammonium concentrations in summer until around 1980 (a factor 3) and then a stabilization or a weak increase. This record is in good agreement with the model results which predict a maximum around 1980–1985 and an increase of a factor of 2.6 between 1920 and 1980, both for calculations using 1997 and 2003 meteorology (Figure 7). The general pattern of past ammonium changes, characterized by a maximum in 1980 followed by a plateau up to 1994, which contrasts with the sulfate trend, can be recognized in the historical NH₃ emission trends (Figure 6). However, emissions from all the countries that make significant contributions to the depositions at CDD increase only by a factor of two. Since the meteorological conditions in the two long-term modeled time series are constant, the higher enhancement of ammonium levels in the model results than in the NH₃ emission input suggests a higher rate of ammonium aerosol formation over the recent decades. Since the production of ammonium aerosols are limited by the availability of sulfate and nitric acid, for which concentrations have increased more than the ammonia emissions up to 1980, a larger proportion of NH₃ is converted to ammonium aerosols in 1980 than in 1925.

[58] For winter, the model calculations suggest an increase of a factor of 2.3 from 1920 to around 1980, somewhat smaller than the corresponding summer trend. The more moderate increase of levels in winter is probably caused by the higher influence of BIC. As described in section 4.2, the BICs prior to 1980 are scaled by the development of NH₃ emissions from USA, which amount to a factor of 2.2 between 1890 and 1980. Since the development of the chemical climate in USA has been similar to that of Europe (with increasingly higher SO₂ and NO_x levels throughout the century and thus a more efficient conversion of NH₃), the trends in the BIC should probably have been higher and the simulated trend of winter level is possibly somewhat underestimated. No obvious winter long-term trends of ammonium is seen in the CDD snow layers (Figure 7). However, the scatter of the individual values are as large as the expected trend, thus a trend cannot not be easily detected. This can also be noted from the model results for the later years when the meteorology for the actual years have been used. For instance, the difference between the modeled values for 2002 and 2003 are almost as large as the trend, despite the fact that the emissions used in the two runs are very similar. Secondly,

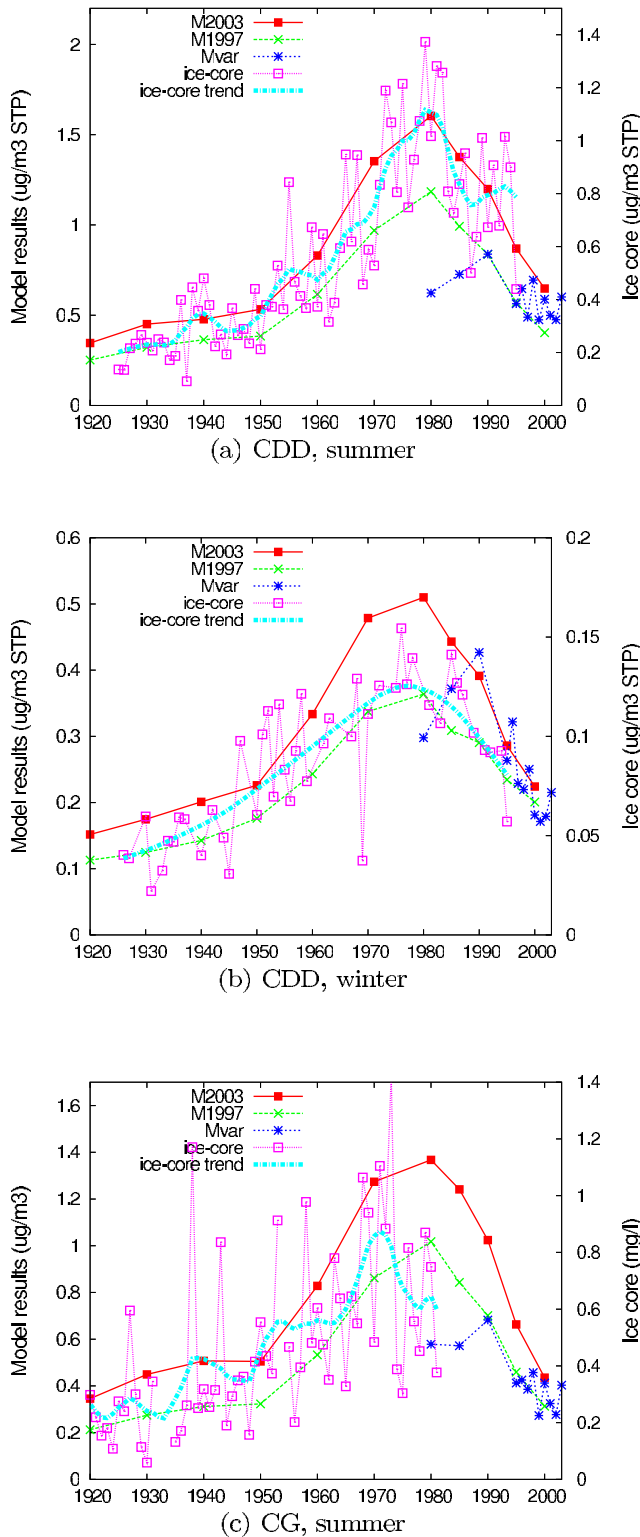


Figure 5. Comparison of modeled trends (model layer 13) and ice core records for sulfate at Col du Dôme (CDD) and Colle Gnifetti (CG). Dashed line with crosses is model simulations using 1997 meteorology (set 2), solid line with solid squares is model simulations using 2003 meteorology (set 3), dashed line with stars is model simulations using emissions and meteorology as appropriate for every year (set 4) dashed line with open squares is ice core record and thick dashed line is smoothed profile (first component of single spectra analysis with a 5-year time window). See section 4.4 for a description of the different sets. The ice core data have been inverted to air concentrations for CDD but not for CG.

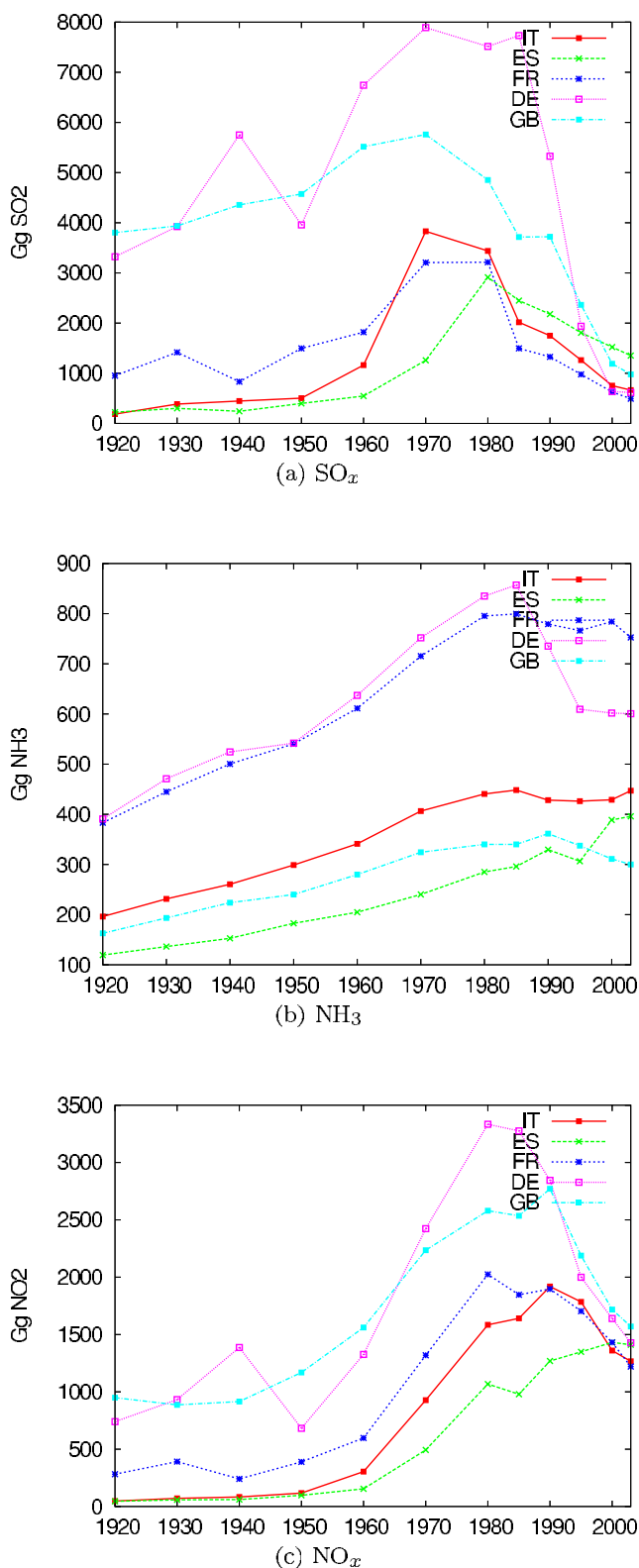


Figure 6. Emissions of SO_2 , NH_3 and NO_x for Italy (IT), Spain (ES), France (FR), Germany (DE) and United Kingdom (GB) for 1920–2003.

the dating of the ice core (e.g., the definition of winter snow layers) is based on the ammonium stratigraphy, e.g., a part of the ice core is defined as “winter” when the ammonium levels are low [Preunkert *et al.*, 2001]. It is possible that this ammonium concentration criteria does not work very well for the more ancient part of the ice core when the ammonium concentrations are lower, and that small parts of winter snow are mixed into the winter layers. For sulfate, however, this would have a weaker effect because of the less pronounced seasonality of sulfate compared to ammonium. An alternative explanation for the increase in ammonium concentrations in the ice core from around 1960 and backward in time is that only a part of the winter snow is accumulated in the deeper part of the CDD ice core. Preunkert *et al.* [2000] found a systematic decrease in the winter to summer snow accumulation with increasing depth in the CDD ice core. The snow in the deeper part of the ice core originates from the slope upstream to the drill site, which is more exposed to wind erosion. It is therefore possible that the dry and cold winter snow is missing in the deeper part of the CDD ice core and that only winter snow from the mild periods, when the snow is more sticky, is collected. The fact that model simulations and the ice core data agree much better for the 1960–1994 period than for the 1925–1960 period indeed suggests partly incomplete winter snow deposition in the lower part of the CDD ice core.

[59] In conclusion, these comparisons between simulated past atmospheric concentrations and inverted CDD ice core records of major inorganic aerosol lead to rather consistent findings for both sulfate and ammonium. This is particularly useful for ammonium since estimates of past NH_3 emissions are a priori far more uncertain than those of SO_2 .

9. Historical Long-Term Trends of Elemental Carbon Aerosol at CDD

[60] Compared to inorganic aerosols like ammonium and sulfate, less data on EC are available for CDD present-day atmospheric concentrations and in the ice core record. Furthermore no investigation of FAR relationship has yet been carried out for EC at the ice core drill site. Present day simulations (2002, albeit with year 2000 emissions) of elemental carbon at CDD give a summer average of $44 \pm 46 \text{ ng m}^{-3}$ STP. This result is in good agreement with the data collected at VO in 2004, which show typical summer values of $40\text{--}50 \text{ ng m}^{-3}$ STP [Legrand *et al.*, 2007]. The model predicts lower levels in winter than in summer ($8 \pm 18 \text{ ng m}^{-3}$ STP) at CDD but no present-day wintertime EC measurement data are available for comparison. As described in section 4.2, free troposphere EC values sampled at Sonnblick were around 20 ng m^{-3} STP. Since our EC boundary conditions are set to zero in these calculations, we probably largely underestimate the winter concentrations at CDD.

[61] Figure 8 shows the calculated trend for CDD and CG for the period 1920–1985 using 2000 meteorology. The absolute concentrations are somewhat higher ($\sim 20\%$) in the simulations where we have used 1997 meteorology, but the trends are similar.

[62] Legrand *et al.* [2007] found that the level of EC in summer CDD layers was enhanced from $4.2 \pm 4.7 \text{ ngC g}^{-1}$

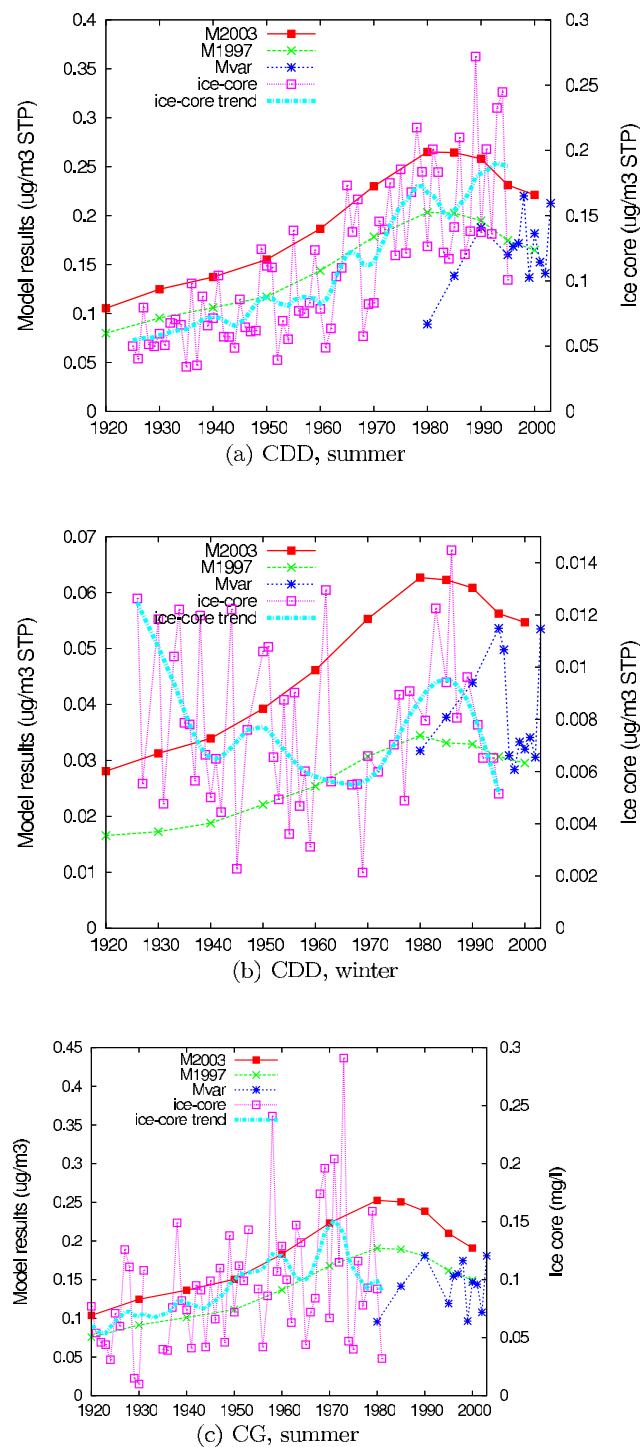


Figure 7. Comparison of modeled trends (model layer 13) and ice core records for NH_x at Col du Dôme (CDD) and Colle Gnifetti (CG). Dashed line with crosses is model simulations using 1997 meteorology (set 2), solid line with solid squares is model simulations using 2003 meteorology (set 3), dashed line with stars is model simulations using emissions and meteorology as appropriate for every year (set 4), dashed line with open squares is ice core record and thick dashed line is smoothed profile (robust spline). See section 4.4 for a description of the different sets. The ice core data have been inverted to air concentrations for CDD but not for CG.

in 1925–1936 to $16 \pm 7 \text{ ngC g}^{-1}$ in 1962–1967, and tends to decrease again to $\sim 10 \text{ ngC g}^{-1}$ in 1989–1991. The EC-C simulations suggest rather unchanged atmospheric concentrations at CDD from 1920 to 1940 followed by a regular increase by a factor of two from 1940 to 1985 (Figure 8). These EC-C simulations therefore differ from the ice record with an absence of maximum around 1970 and a weaker long-term change. However, the assumptions made concerning variable emission factors for road traffic emissions (section 3.2) have a very strong effect on the simulated trends, especially for summer. The model simulations with the EC-V emission estimates predict an increase in EC levels of about a factor of 4–5 between 1920 and 1970, giving a better match between the ice core and modeled data (Figure 8). The latter results suggest that the EC-V emission estimates are more realistic, but they also illustrate the large uncertainties involved in these calculations.

[63] As seen in Figure 8, the simulated summer and winter trend at CDD using EC-V emissions are very different with a much lower increase in winter (a factor of 2 to 3) than in summer (a factor of 4 to 5) between 1920 and 1970. This difference is caused by the larger increase of mobile source emissions (S7 and S8) compared to the emissions from stationary sources since stationary combustion sources dominates in wintertime whilst the emissions in summer originates predominantly from mobile sources. Investigations of EC in winter CDD layers were limited to a few samples, but a similar level was found in 1982–1984 and 1937–1939 (2.4 ngC g^{-1} and 3.8 ngC g^{-1} , respectively) whereas the level in 1973–1976 was slightly higher (6.5 ngC g^{-1}) [Legrand *et al.*, 2007]. Although further studies are needed here to confirm such a more moderate long-term change in winter with respect to summer in Alpine ice core record, this is consistent with the model simulations. As discussed above, the assumption of zero EC boundary conditions is a major uncertainty in the calculation of winter EC concentrations at CDD. However, the consistency between the simulated and the ice core trend indicates that either the background levels have changed with the same trend as the influence from European sources, or, less likely, that the background level (boundary conditions) is a negligible source for the EC sampled at CDD.

[64] Another uncertainty in our model simulation for EC trends at CDD is the lack of inclusion of biomass burning in our emission inventories. A number of studies have shown that forest fire emissions from North America can have significant impacts over 1000s of km, including over Europe [Forster *et al.*, 2001; Simmonds *et al.*, 2005; Stohl *et al.*, 2006]. Indeed, during the extreme forest fire episode of August 1998, Forster *et al.* [2001] showed that Canadian forest fire emissions accounted for almost 60% of the enhancements in CO seen at the background sites of Mace Head, Ireland. Enhancements in black carbon at Mace Head were lower than those of CO (because of the greater washout of BC), but still significant compared to normal background concentrations. However, these papers have largely dealt with specific episodes, so do not allow a direct estimate of the OBB contribution to measurements at CDD. As discussed in section 3.2, Gelencsér *et al.* [2007] found EC from biomass burning to be minor compared to EC from fossil fuel for high-elevated sites at present day, especially in summer.

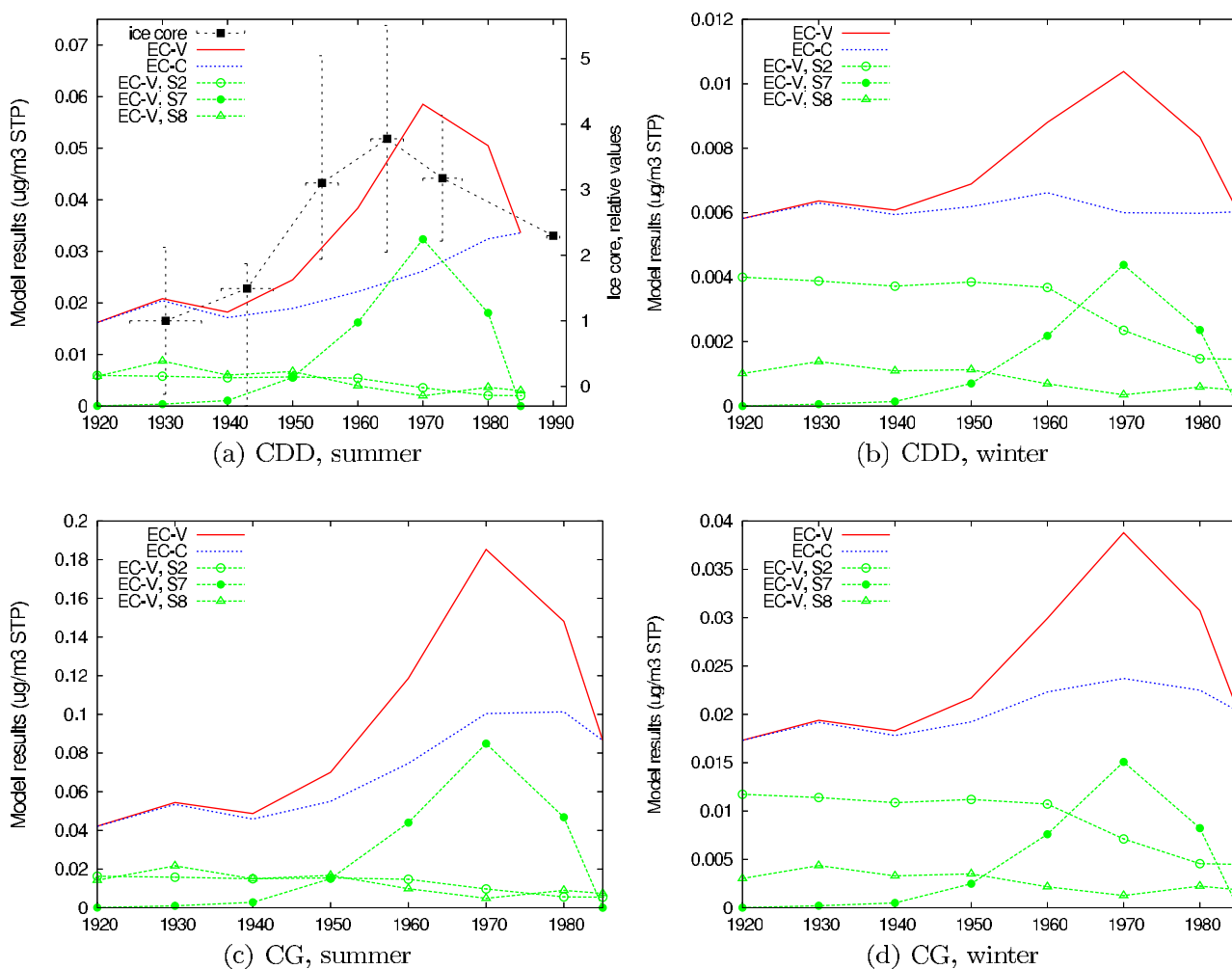


Figure 8. Modeled trends at Col du Dome (CDD) and Colle Gnifetti (CG), model layers 13, year 2000 meteorology. Solid line is total EC with varying emission factors (EC-V). Dotted line is EC with constant emission factors (EC-C). Dotted lines with marks show contributions from domestic emissions (open circles), road traffic (sector S7, with variable emission factors, solid circles), and from other mobile sources (sector S8, triangles). For CDD summer, ice core data are given relative to ~ 1930 (black, dotted line with solid squares). See *Legrand et al.* [2007] for absolute numbers and a detailed discussion of these ice core samples.

[65] Although estimates exist of the historical trends in anthropogenic emissions, including domestic biomass burning [*Ito and Penner, 2005*], it is harder to assess the changes in the contribution from wildfires to the trends over this period. A number of results suggest, however, that forest fire emissions have not contributed significantly to the observed trends of aerosol at CDD during the 20th century. Results presented by *Holdsworth et al.* [1996], based upon examination of NH_4^+ , black carbon and concentrations of insoluble microparticles at ice cores in Greenland and Canada, suggest that the major period of biomass burning in North America occurred in the last half of the 19th century. Concentrations of all of these “marker” species (albeit imperfect) show little change from around 1920 to the end of the century.

[66] Thus, although forest fires may make significant contributions on an episodic basis to concentrations over Europe and likely the Alpine region, most likely neither

European nor American OBB sources are responsible for the observed trends at CDD.

10. Comparisons With Another Alpine Ice Core at CG

[67] The source-receptor relationships presented in section 7 indicate that in summer Spain, France, Italy and Germany account together for 50% of total sulfate at both CDD (Figure 4) and CG (not shown). There are however some differences when moving eastward from CDD to CG; the Spanish and French contribution decrease but are compensated by increasing contributions from Germany and Italy. From Figure 6 we see that emissions from Germany and France were relatively constant between 1970 and 1980, whilst Spanish emissions maximized in 1980 and those from Italy in 1970. As a result the model predicts a maximum in summer sulfate levels in 1980 at both CDD and CG but the

increase between 1970 and 1980 is less pronounced at CG than at CDD.

[68] In Figure 5 we compare model simulations with the sulfate CG ice record. As seen from Figure 5, simulations and ice record are in reasonable agreement from 1920 to 1970. In the CG ice cores a rather sudden decrease of concentrations is observed after 1970. One reason for this difference with respect to CDD may of course be the lower influence of Spanish emissions at this site. However, there may be some differences in behavior of the snow packs also. At CG summer snow is mainly preserved whereas the dry snow in winter is strongly eroded by wind. It is possible that after 1970 the CG snow layers start to contain a small amount of winter snow, causing lower concentrations in the snow packs. Note that, when a careful seasonal dissection is applied to recent CG snow deposits, a good agreement is found for sulfate level for both summer and winter [Preunkert *et al.*, 2000].

[69] In contrast to the observations for sulfate, there is a large difference in elemental carbon concentrations simulated at CDD compared to CG (see section 9), the latter having concentrations some 5 times higher in summer (Figure 8). This difference in simulations arises from the rather large EC emissions from the Po valley in the vicinity of the CG site. As noted in section 3.2, summertime EC emissions are usually dominated by road traffic sources, and in the EMEP grids (including Milan) just northeast of CG emissions reach up to $\sim 500 \text{ kg km}^{-2} \text{ yr}^{-1}$. Emissions on the French side of the Alps and around CDD are typically factors of 2–10 lower than this. As discussed by Legrand *et al.* [2007], ice core concentrations of EC are also almost 6 times higher at CG than at CDD in summer ice. The methods deployed to gain these ice core data were different at the two sites and can possibly lead to a difference by a factor of two at the best. Our simulations point out that this large difference between the two sites is more likely to be caused by the different impact from the emissions in the Po Valley.

11. Conclusions

[70] Previous studies of Alpine ice cores have relied on emission estimates available for annual timescales, and have had to make assumptions concerning the countries likely to contribute to the ice core trends. In this study the use of the EMEP chemical transport model allows for a more systematic evaluation of the sources contributing to a given site, and takes into account differences in both emissions and meteorological factors over the years. Furthermore, the model can account for nonlinear changes in the emission-deposition relationships, for instance due to changes in the oxidative capacity of the atmosphere or in the equilibrium between ammonium, sulfate and nitrate aerosols and nitric acid, sulfuric acid and gaseous ammonia.

[71] In summertime, the high Alpine CDD site lies within the boundary layer, and is subject mainly to European influences. In wintertime the site is usually above the boundary layer, and the influence of sources from outside our model domain is much larger, in particular for sulfate and most likely also for EC. The winter simulations for sulfate are to a large extent tied to the winter ice core values, as these are used to scale the inflow from the western boundary. In summer, the larger contributions from Euro-

pean sources give rise to a different trend with a greater increase of sulfate, especially in the 1960–1980 period, in good agreement with the ice core record.

[72] Both the NH_x trend derived from the summer CDD record and the simulated summer trend show a higher enhancement of levels between 1920 and 1980 (\sim factor of 3) than the ammonia emissions in surrounding areas (\sim factor of 2). The model simulations indicate that these nonlinearities in the emission-deposition relationship can be explained by a faster conversion of gaseous ammonia to ammonium aerosols caused by higher availability of sulfuric acid and nitric acid resulting from the large increase of SO₂ and NO_x emissions during the last century.

[73] For EC, emissions in summertime are dominated by mobile sources. Therefore summer trends in the historical development of EC are driven much more by the emission trends for this sector than by trends in the stationary combustion sources. The combination of decreasing historical emission factors for vehicles and increasing road traffic leads to a maximum in EC emissions for road transport around 1970, which is reflected in the simulated EC trend and also in agreement with the ice core record.

[74] For the inorganic aerosols sulfate and ammonium the absolute levels, as well as the trends, are rather similar at CDD and CG. For elemental carbon, the model simulations demonstrate that the considerably higher concentrations (\sim factor 6) in the CG summer ice compared to CDD at least partly can be explained by the vicinity of the CG site to the large sources in the Po Valley.

[75] Despite the large uncertainties in the emission data, especially for EC but also for NH₃, the agreement between model simulations and the ice core records is rather encouraging and indicates that the historical emission inventories are reasonable. This study also shows that the combination of historical inventories and modeling is a valuable tool when interpreting ice core trends and comparing data from different sites.

[76] **Acknowledgments.** This work was supported by the European Commission's Fifth Framework Program in the form of the CARBOSOL Project (contract EVK2-2001-00067), as well as by the Cooperative Programme for Monitoring and Evaluation of the Long-Range Transmission of Air Pollutants in Europe (EMEP) under UNECE, and it is a contribution to the ACCENT Network of Excellence under the European Commission.

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Direct shortwave radiative forcing of sulfate aerosol over Europe from 1900 to 2000

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[1] On the basis of historical simulations of the atmospheric distribution of sulfate aerosol over Europe, we have estimated the evolution of the direct shortwave radiative forcing due to sulfate aerosol from 1900 to the present day. Following the trend of atmospheric sulfate burden, the radiative forcing reaches its peak in the 1980s. Since then, environmental policies regulating SO_x emissions successfully reduced the atmospheric load. On average, the forcing of the year 2000, representing present day, equals that of the 1950s. Spatially, the forcing maxima experienced a shift from the northwest to the southeast during the century. The ship emissions of sulfur keep increasing since the 1980s, hence their relative contribution to the sulfate load and radiative forcing constantly increased, from 3% in the 1980s to over 10% in the year 2000. Forcing is strongest during summertime, with a seasonal mean of -2.7 W m^{-2} in the 1980s and -1.2 W m^{-2} in summer 2000. The mean forcing efficiency is slightly reduced from $-246 \text{ W (g sulfate)}^{-1}$ in the 1900s to $-230 \text{ W (g sulfate)}^{-1}$ in the year 2000, and it declines with changed geographical distribution of sulfur emissions.

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1. Introduction

[2] Anthropogenic emissions have caused serious environmental problems in Europe since the beginning of industrialization, contributing to soil, water and air pollution. Air pollution affects climate through absorbing and scattering aerosol particles, which can warm or cool the Earth-atmosphere system. Emissions of SO_x and black carbon are particularly relevant for the climate.

[3] SO_x , emitted mostly as SO_2 gas, is converted in the atmosphere via gaseous and aqueous chemical reaction to sulfate aerosol. Sulfate aerosol has an impact on climate via direct and indirect radiative forcing. For sulfate aerosol, the direct effect results from scattering of incoming solar radiation back to space. The indirect effect results from the ability of the sulfate aerosol particles to act as cloud condensation nuclei, resulting in more but smaller cloud droplets, increasing the cloud albedo [Twomey, 1974]. On the other hand, smaller cloud droplets can suppress precipitation, prolonging the life time of the cloud and the aerosol [Albrecht, 1989]. The combination of these effects and their feedbacks is up to now poorly understood [Intergovernmental Panel on Climate Change (IPCC), 2001a].

[4] Additional uncertainties exist about the direct, semi-direct and indirect effects of black carbon. Black carbon strongly absorbs the solar radiation and warms the aerosol layer. It can trap more energy than is lost by scattering to space to overall warm the Earth-atmosphere system. Absorption of solar radiation by black carbon particles in clouds can evaporate cloud droplets reducing cloud cover [Ackerman *et al.*, 2000]. Further, black carbon particles in cloud droplets can reduce the cloud albedo [Krüger and Grassl, 2002] and can act as ice nuclei [DeMott *et al.*, 1999].

[5] Considering the climate impact of both sulfate and black carbon aerosols presents yet another problem: for newly released particles their climate impact can be treated individually, whereas aged particles are internally mixed. Internally mixed particles have different optical and hygroscopic properties depending on their age and chemical composition [Lesins *et al.*, 2002]. Haywood *et al.* [1997], Myhre *et al.* [1998] and Lesins *et al.* [2002] showed that an internal mixture of sulfate and black carbon results in substantially different forcing than an external mixture.

[6] In this study we investigate the historical evolution of the aerosol radiative forcing over Europe during the 20th century. We look at the direct forcing only, because it is the best understood aerosol effect. The study is further limited to sulfate aerosol, since a consistent historical emission inventory of black carbon in Europe in suitable resolution is not yet available.

[7] Myhre *et al.* [2001] estimated the averaged global radiative forcing evolution from 1750 to 1995, utilizing a radiation transfer model. All known radiative forcings, greenhouse gases, ozone, anthropogenic and natural aerosols (including sulfate and black carbon), and solar irradi-

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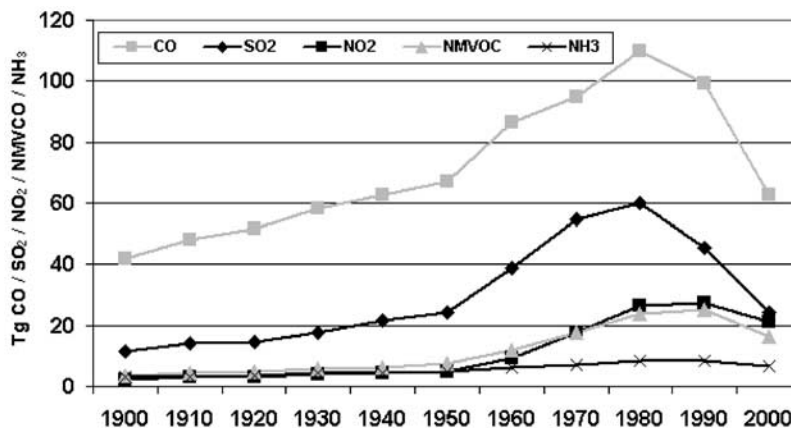


Figure 1. European emissions included in the EMEP domain (CO, SO₂, NO₂, NMVOC, NH₃, natural and anthropogenic (Tg/Year)).

ance radiation, were considered. The global mean direct aerosol forcing was computed assuming internal and external aerosol mixtures. The evolution of the sulfate aerosol forcing was calculated scaling to global mean SO₂ emissions. This study points out the increasing importance of the anthropogenic aerosol forcing for the past 100 years. The global mean direct radiative forcing of sulfate of -0.05 W m^{-2} is given for the 1900s, its value doubled in 1945, almost constantly increasing to -0.36 W m^{-2} by 1995, with only a minor decrease during the 1980s.

[8] *Boucher and Pham* [2002] computed the direct and indirect sulfate aerosol forcing over the period 1850 to 1990 applying a global GCM. They focused on sulfate aerosol only, because more information was available for sulfur sources than for other aerosol species. They found that the global direct sulfate forcing has increased from near zero [1850] to -0.42 W m^{-2} in 1990, with nearly constant forcing efficiency of $-150 \text{ W}(\text{g sulfate})^{-1}$ (ratio of the radiative forcing to the anthropogenic sulfate burden). The global forcing was found to be fairly constant between 1980 and 1990 because of emissions reduction in the US and Europe, with a spacial shift from the US, Europe, Russia and the North Atlantic to Southeast Asia and the Indian and Pacific Oceans.

[9] Another study on the trend in tropospheric aerosol loads and the direct radiative forcing was carried out by *Tegen et al.* [2000]. The global trend of the atmospheric load and forcing of sulfate and carbonaceous aerosols from fossil fuel burning as well as soil, dust and sea salt from 1950 to 1990 was constructed. In 1950, the global averaged optical thickness of anthropogenic sulfate aerosol (0.005) was found to be smaller than that of natural sulfate (0.007), it more than doubled by 1990s (0.011). Globally, a range of top-of-atmosphere direct forcing of -0.5 to $+0.1 \text{ W m}^{-2}$ was found because of the uncertainty in the contribution of the black carbon aerosol.

[10] For our study we have utilized a regional model, which provides a better resolution than the earlier performed global simulations, allowing us to have a closer look at different regions in Europe. In order to better demonstrate the regional differences, we have calculated sulfate direct forcing for five selected areas in Europe, representing different geographical and atmospheric conditions, and discuss

the different trends for each of these areas. Ship emissions is the only constantly growing source of sulfur in Europe. A sensitivity study without ship emissions investigates the trend of their relative contribution to the direct radiative forcing during the past century. The results of the present study serve as a baseline for future investigations on aerosol climate impact evolution over Europe, which should include black carbon.

2. Emissions

[11] In this section, we describe the emission data used in the assessments with the EMEP Unified model [*Simpson et al.*, 2003a] discussed in the next chapter. The emission data has been collected from different sources depending on the availability of emission years, compounds, sectoral breakdown, resolution and quality considerations. The emissions are depicted on the basis of emission levels and sector distribution, spatial and temporal distribution and information about natural and shipping emissions (Figure 1 and Table 1).

2.1. Emission Levels

[12] The anthropogenic emission input data of SO₂, NO_x, NH₃, CO and NMVOC from 1980 to 2003 are based as far as possible upon emissions reported per sector and grid officially reported to the Convention on Long-Range Transboundary Air Pollution [*Vestreng et al.*, 2004].

[13] For the period prior to 1980, we have used three different sources of information (Table 2) in consistency with EMEP data for the years 1980–2000.

[14] An evaluation of the *van Aardenne et al.* [2001] inventory for SO₂ reveals that this inventory has rather a constant level of emissions between 1980 and 1990, while *Lefohn et al.* [1999] show an increase of European sulfur emissions in the 1980s. We have compared the *van Aardenne et al.* [2001] and *Mylona* [1996] emission inventories (excluding the former USSR/CIS emission estimates), and find that there are substantial differences in emission levels and trends particularly from 1950 onward. *Mylona* [1996] emissions per country show close agreement with the EMEP official emissions which have been validated by models and measurements between 1980–2000 (section 3.2), hence *Mylona* [1996] was our preferred choice for SO₂ inventory.

Table 1. European Emissions Included in the EMEP Domain, Natural and Anthropogenic^a

| Year | SO _x (TgSO ₂) | | | NO _x (TgNO ₂) | | | NMVOC | | | NH ₃ | | | CO | | |
|------|--------------------------------------|-------|--------------------|--------------------------------------|-------|--------------------|-------|-------|--------------------|-----------------|-------|--------------------|--------|-------|--------------------|
| | Land | Ships | Other ^b | Land | Ships | Other ^b | Land | Ships | Other ^b | Land | Ships | Other ^b | Land | Ships | Other ^b |
| 2000 | 17.36 | 2.72 | 4.16 | 16.95 | 3.85 | 0.32 | 16.01 | 0.081 | 0.39 | 6.06 | - | 0.56 | 61.08 | 0.33 | 1.25 |
| 1990 | 39.19 | 2.13 | 4.16 | 23.84 | 3.01 | 0.32 | 24.55 | 0.081 | 0.39 | 7.92 | - | 0.56 | 97.55 | 0.33 | 1.25 |
| 1980 | 54.16 | 1.66 | 4.16 | 23.81 | 2.35 | 0.32 | 23.47 | 0.050 | 0.40 | 8.04 | - | 0.56 | 108.13 | 0.20 | 1.31 |
| 1970 | 50.29 | 1.02 | 3.59 | 15.85 | 1.44 | 0.23 | 17.24 | 0.030 | 0.40 | 6.79 | - | 0.43 | 93.43 | 0.12 | 1.11 |
| 1960 | 34.93 | 0.57 | 3.23 | 8.07 | 0.81 | 0.18 | 11.99 | 0.017 | 0.14 | 5.74 | - | 0.33 | 85.98 | 0.07 | 0.94 |
| 1950 | 21.10 | 0.32 | 3.03 | 4.37 | 0.45 | 0.15 | 7.26 | 0.009 | 0.09 | 4.77 | - | 0.29 | 66.12 | 0.04 | 0.75 |
| 1940 | 18.38 | 0.36 | 2.97 | 4.10 | 0.33 | 0.13 | 6.18 | 0.007 | 0.07 | 4.09 | - | 0.18 | 62.00 | 0.03 | 0.64 |
| 1930 | 14.37 | 0.29 | 2.90 | 3.35 | 0.41 | 0.13 | 5.47 | 0.009 | 0.06 | 4.05 | - | 0.16 | 57.77 | 0.04 | 0.52 |
| 1920 | 11.59 | 0.18 | 2.88 | 2.86 | 0.26 | 0.12 | 4.61 | 0.005 | 0.05 | 3.30 | - | 0.12 | 50.99 | 0.02 | 0.44 |
| 1910 | 10.92 | 0.19 | 2.87 | 2.60 | 0.27 | 0.11 | 4.17 | 0.006 | 0.05 | 2.78 | - | 0.10 | 47.79 | 0.02 | 0.38 |
| 1900 | 8.45 | 0.12 | 2.86 | 2.09 | 0.17 | 0.11 | 3.46 | 0.00 | 0.04 | 3.10 | - | 0.09 | 41.52 | 0.02 | 0.33 |

^aUnit is Tg/Year.

^bOther, emissions from North African and Asian areas within the EMEP domain. In addition natural emissions (volcanoes, approximately 2 Tg/Yr SO₂, and DMS, 0.8 Tg/Yr SO₂) are included for SO_x.

Berglen et al. [2007] concluded that model assessments performed with the EMEP inventory better reproduced the trends in observations 1985–2000 than the *Smith et al.* [2004] inventory. Emissions available in the EMEP database (<http://webdab.emep.int/>, and V. Vestreng and A. Semb, Nitrogen oxides emission inventories over Europe since the preindustrial era, unpublished manuscript, 2006, hereinafter referred to as Vestreng and Semb, unpublished manuscript, 2006) show that the land based European sector emissions have been continuously reduced between 1980 and 1990, in accordance with *Mylona* [1996]. Thus we have relied on the European SO₂ inventory developed by *Mylona* [1996] despite that this inventory originally lacks the sectoral breakdown.

[15] The SO₂ inventory from *Mylona* [1996] were distributed according to EMEP 1980 sector distribution for SNAP (Selected Nomenclature for Air Pollutants, http://reports.eea.eu.int/EMEP_CORINAIR3/en/) for the sectors 1, “Combustion in energy and transformation industries”; 2, “Nonindustrial combustion plants”; 3, “Combustion in manufacturing industries;” and 4, “Production processes,” while sector 7, “Road transport,” and 8, “Other mobile sources and machinery,” are scaled according to the petroleum consumption.

[16] The NO_x inventory included is developed by Vestreng and Semb (unpublished manuscript, 2006) by country and SNAP sector. This regionally developed NO_x inventory compares much better with Alpine ice core data than the emissions from *van Aardenne et al.* [2001]. In particular the trend from 1950 to 1980 is much steeper in the Vestreng and Semb (unpublished manuscript, 2006) inventory as a result of the variable emission factors for road transport included.

[17] For NH₃, NMVOC and CO, we have included the *van Aardenne et al.* [2001]. In the case of CO and NMVOC there were to our knowledge no other inventories published. A historical emission inventory of ammonia including 12%

natural emissions per European country for the years 1870, 1920, 1950 and 1980 has been provided by *Asman and Drukker* [1987]. Their anthropogenic emission inventory is comparable to that of *van Aardenne et al.* [2001] between 1950 and 1980 but estimates by *Asman and Drukker* [1987] exceed those of *van Aardenne et al.* [2001] before 1950, e.g., they estimate 20% more emissions in 1920 in Europe. Our choice of the *van Aardenne et al.* [2001] emission inventory for NH₃ is due both to their more extensive database (more years included), and because their 1980 data compares better with officially reported data for Europe [*Vestreng et al.*, 2004].

[18] The *van Aardenne et al.* [2001] data is available in a 1° × 1° degree grid and have been adapted onto SNAP source categories according to the EMEP 1980 sector split.

[19] The ships emissions of SO₂, NO_x, NMVOC and CO in the period from 1880 to 1975 have been assumed to be directly proportional to the registered tons associated with the steam and motor ships as detailed in *Mitchell* [1981]. For the years 1990 and 2000 emission estimates from *Lloyd's Register of Shipping* [1995, 1998, 1999] and from ENTEC [*Whall et al.*, 2002], respectively, are available. We have used the year 2000 emissions to downscale the ships emissions back to 1980, assuming a growth rate of 2.5% per year, or a factor 1.28 per decade [*Endresen et al.*, 2003].

[20] In addition, natural emissions of SO₂ from volcanoes and ocean phytoplankton (DMS) are included. The emissions from volcanoes are constant (2 Tg) throughout the period 1900–2000, equal to the Italian reported natural emissions of sulfur released from Mount Etna [*Vestreng et al.*, 2004]. The DMS inventory is constant over the years [*Tarrason et al.*, 1995].

[21] Emissions from areas included in the EMEP domain but outside Europe (i.e., North African and some Asian countries), are from *van Aardenne et al.* [2001].

Table 2. Emission Data Characteristics for Inventories Included in This Study

| Emission Data Source | Pollutant | Years | Sectors | Resolution |
|---|---|-----------|---------|--|
| <i>Mylona</i> [1996] | SO ₂ | 1880–1990 | - | country/Europe/5 years interval |
| VestrengSemb (unpublished manuscript, 2006) | NO _x | 1880–1985 | SNAP | country/Europe/5 years interval |
| <i>van Aardenne et al.</i> [2001] | NH ₃ , VOC, CO | 1880–1990 | EDGAR | 1° × 1° global/10 years interval |
| <i>Vestreng et al.</i> , 2004 | SO ₂ , NO _x , NH ₃ , NMVOC, CO | 1980–2003 | SNAP | 50 × 50 km ² /Europe/annually |

2.2. Emission Distribution

[22] The temporal distribution of emissions has been done according to the Generation and Evaluation of Emission Data project (GENEMIS [Friedrich and Reis, 2004]), a subproject of EUROTRAC-2. (EUROTRAC-2 is the EUREKA Project on the Transport and Chemical Transformation of Environmentally Relevant Trace Constituents in the Troposphere over Europe; Second Phase (<http://www.gsf.de/eurotrac/>.) Monthly and daily factors are applied for the temporal emission distribution. The factors are specific to each pollutant, emission sector and county, taking into account the different climates and hence energy use across Europe.

[23] The heights of the stacks have changed significantly during the last century, which we have taken into account by defining a “tall stack” (1955–present) and a “low stack” (previous to 1955) period. During 1880–1945, stack heights were typical about 40 meters to “clear the roof tops and neighboring house” [Brimblecombe, 1987]. The stack heights increased to around 150 m in 1950–1975 as a result of increasing environmental concern (“tall stack policy”). In the model calculations, the effective emission heights of power plants and industry are moved one model layer closer to the ground in the low-stack period in an attempt to reproduce the historical distribution of stack heights. In both cases, the emissions rapidly become well mixed in the boundary layer and the effect is only minor.

[24] The emissions were distributed horizontally the following way. The *van Aardenne et al.* [2001] inventories were available per country with the present country borders. Scaling factors per country and sector were deduced by

$$f_{\text{Year}}(\text{country, sector}) = \frac{\text{emission}_{\text{Year}}(\text{country, sector})}{\text{emission}_{1980}(\text{country, sector})}$$

and used to scale the EMEP 1980 emissions backward in time, but ensuring that the total emission fractions were kept. In this way, the finer resolution ($50 \times 50 \text{ km}^2$) of the EMEP data could be kept and the evolution of the historic emissions included. In the data from *van Aardenne et al.* [2001], spatial distribution over the years is only different when the relative amount within the sectors change. Thus we lose no information when applying only the scaling factors as the sector information is kept.

[25] For SO_2 and NO_x , emission scaling factors were defined in the same way as for NH_3 , VOC and CO. The historical emissions for SO_2 and NO_x were available from 1880–1985 per country with country borders as they were historically. For instance, emissions are not available separately for the countries within the Former Soviet Union, and emissions from Austria did include emissions from the whole Austrian Empire before 1920. Thus we have made the following assumptions: The countries in the Former Soviet Union are scaled with the same factors from 1920 to 1980. East and West Germany are scaled separately back to 1950, but prior to this as a sum. The areas corresponding to Czech Republic and Slovakia are scaled with emissions for Former Czechoslovakia back to 1920. In the same period, Slovenia, Croatia, Bosnia and Herzegovina, Serbia and Montenegro and The Former Yugoslav Republic of Macedonia are scaled using the historic emissions of Former Yugoslavia. For the years prior to 1920, the changes of

borders have been extensive and more crude assumption had to be made. Thus the uncertainties related to spatial distribution in this period are larger. Austria was a part of the Austrian Empire, including Czech Republic and Slovenia. For, e.g., 1910, Austria is scaled with

$$\frac{\text{Austrianemissions}(1920)}{\text{Austrianemissions}(1980)} \times \frac{\text{AustrianEmpireemissions}(1910)}{\text{AustrianEmpireemissions}(1920)},$$

where the Austrian Empire emissions for 1920 are estimated as the sum of emissions from Austria plus $X \times$ Former Czechoslovakia and $Y \times$ Former Yugoslavia. X has been defined as Czech Republic/(Czech Republic+Slovakia) for 1980, Y as Slovenia/(Slovenia, Croatia, Bosnia and Herzegovina, Serbia and Montenegro and The Former Yugoslav Republic of Macedonia) for 1980. X and Y have been defined separately for SO_2 and NO_x emissions. The same procedure has been followed to define scaling factors for Hungary prior to 1920, as The Hungarian Kingdom included Hungary, Slovakia and Bosnia and Herzegovina. Before 1920, Russia included Poland, and United Kingdom included Ireland. Similar procedures were followed for these countries.

3. Model Setup

3.1. Unified EMEP Model

[26] The Eulerian EMEP model [Simpson *et al.*, 2003a] was utilized to determine the historical sulfate aerosol distribution. It is a multilayer atmospheric dispersion model designed for simulating the long-range transport of air pollution over several years. The model domain is centered over Europe and also includes most of the North Atlantic and the polar region. The model has 20 vertical layers in σ -coordinates below 100hPa. It is primarily intended for use with a horizontal resolution of $\sim 50 \times 50 \text{ km}^2$ (at 60°N) in the EMEP polar stereographic grid. The chemical scheme uses about 140 reactions between 70 species. SO_2 is oxidized to sulfate both in gas phase with OH and in aqueous phase through H_2O_2 , O_3 and O_2 catalyzed by metal ions, assuming a fixed pH value of 4.5. The sulfur chemistry is coupled to the photochemistry, thus changes in the oxidation capacity may change the SO_2 oxidation rate and vice versa. The partitioning between ammonia, ammonium, sulfate, nitrate and nitric acid is calculated using the EQSAM module detailed by Metzger *et al.* [2002a, 2002b].

[27] The dry deposition module makes use of a stomatal conductance algorithm which was originally developed for calculation of ozone fluxes, but which is now applied to all pollutants where stomatal control is important [e.g., Simpson *et al.*, 2003b; Tuovinen *et al.*, 2004]. Dry deposition of aerosol particles depends on their size, with the model version used here distinguishing between fine and coarse aerosols. Details of the formulation are given by Simpson *et al.* [2003a]. Parameterization of the wet deposition processes in the EMEP model includes both in-cloud and subcloud scavenging of gases and particles, using scavenging coefficients.

[28] The EMEP Unified model use meteorological data from PARLAM [Benedictow, 2002], a dedicated version of the operational HIRLAM model (High Resolution Limited Area Model) maintained and verified at Norwegian Mete-

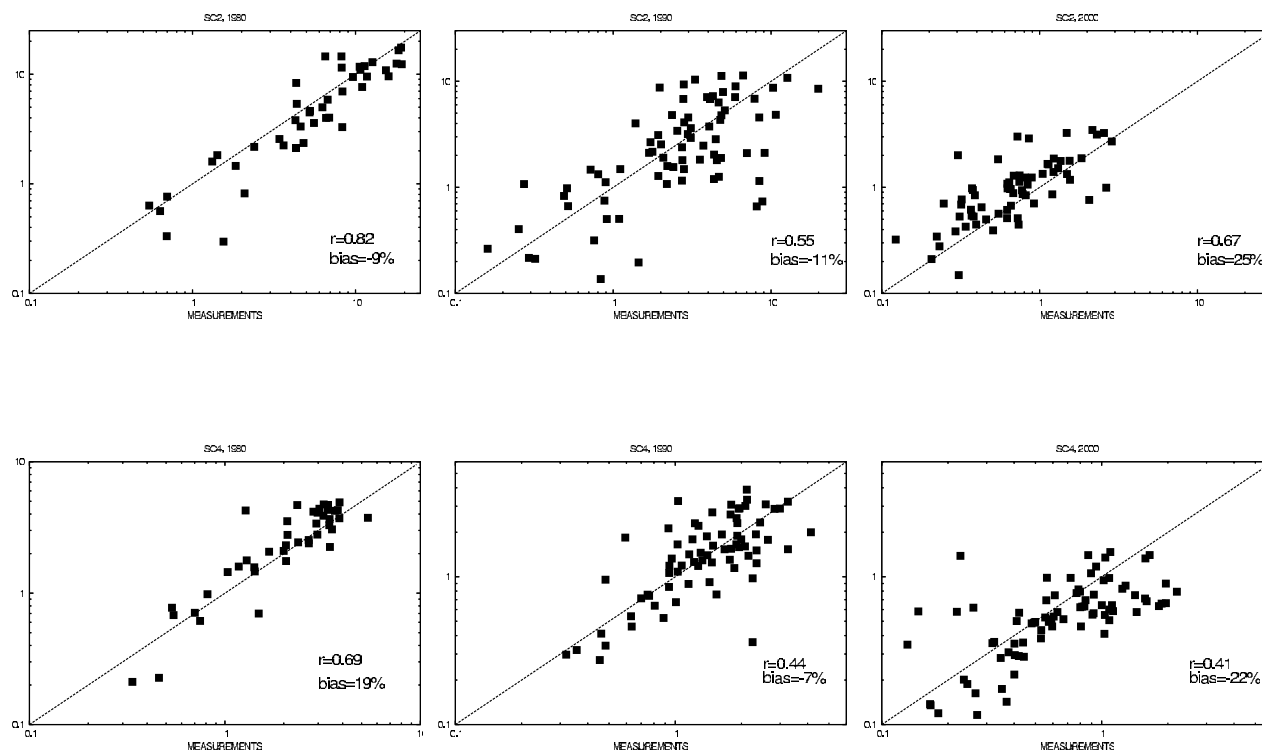


Figure 2. Scatterplots of EMEP model results versus measurements for (top) sulfur dioxide (ppbv) and (bottom) sulfate in air ($\mu\text{g}(\text{S})\text{m}^3$) for 1980, 1990 and 2000.

orological Institute. The numerical solution of the advection term is based upon the fourth-order Bott scheme [Bott, 1989a, 1989b]. The lateral boundary conditions for most species are based on measurements as described by Simpson *et al.* [2003a] and Fagerli *et al.* [2004]. Furthermore, a scaling factor has been applied to account for the change in the concentrations at the boundary during the time period 1900–2000. This factor has been defined on the basis of the EPA emissions (U.S. Environmental Protection Agency [2000], and updates on their web page, <http://www.epa.gov>) for SO_x and NO_x emissions for the 1980–2000 period. The trends in ammonium aerosol were set by weighting the trend of SO_x emissions with 2/3 and NO_x emissions with 1/3, assuming that the production of ammonium nitrate and ammonium sulfate determines the trend. For the period prior to 1980, we have used the winter trends of sulfate and nitrate from an ice core at Col du Dome (4250 m asl, French Alps). In the winter time, Col du Dome is above the boundary layer most of the time. Most of the air pollution deposited at this site originates from sources outside the western boundary of the EMEP domain [Fagerli *et al.*, 2007], thus the trend extracted from the ice core is a good indicator of the air pollution arriving from North America. For ammonium, we used the trends of NH_3 emissions from United States derived from [van Aardenne *et al.*, 2001].

[29] For this study, the EMEP model is run for every tenth year for the period 1900–1980 plus 1985, 1990, 1995 and 2000 using appropriate boundary conditions and emissions. The meteorological year is 1997 for all the model runs. This was done because no set of meteorological data back to 1900 was available. Consequently, the interannual variability

which has an important influence on sulfate aerosol distribution and its forcing could not be considered.

3.2. EMEP Model Evaluation

[30] Sulfur dioxide and sulfate background concentrations have been monitored in Europe at several sites since around 1980. Here, we briefly present model evaluation against SO_2 and SO_4^{2-} measurements at the EMEP monitoring sites. For this evaluation we use the same model version revision 2.0, except that for the evaluated version the meteorology for the specific years has been applied (unlike in this study). Because of scarcity of continuous measurements from 1980 to 2000, and in order to evaluate the model's ability to reproduce the changes in sulfur levels over as large parts of Europe as possible, we use all the EMEP observations available in 1980, 1990 and 2000, respectively. Figure 2 shows scatterplots of model results versus observations for yearly averages of all the available EMEP sites measurements of sulfur dioxide and sulfate. The model tends to over predict the levels of sulfur dioxide somewhat in the recent years (bias = 25%), whilst in the beginning of the period the sulfur levels are underestimated by 9% on average. The opposite can be noted for sulfate, with model results 19% higher than the observations in 1980, and 22% lower in 2000. Sulfate in precipitation (not shown) behaves similar to sulfate in air. These results might indicate a too high SO_2 to SO_4^{2-} oxidation rate in the beginning of the period and a too low oxidation rate in the end. However, there are other possible explanations. In general, the quality of both the emissions and the measurements have become better in recent years [e.g., Vestreng *et*

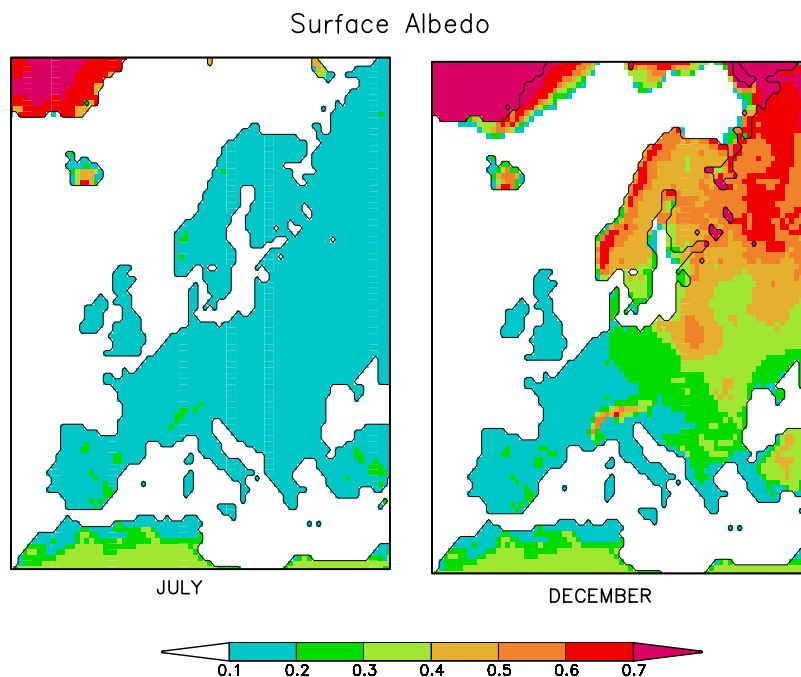


Figure 3. Mean modeled surface albedo for July and December 1997.

al., 2007]. Moreover, the wet scavenging process is complicated and difficult to represent in atmospheric models, partly because of the uncertainty related to prediction of precipitation. In addition, the deposition of atmospheric SO_2 to plant surfaces stimulate the deposition of ammonia and vice versa, because of neutralizing effects [e.g., *van Hove et al.*, 1989; *Flecharde et al.*, 1999]. The marked decrease in SO_2 emissions combined with small changes in NH_3 emissions implies a generally higher present-day dry deposition of SO_2 than at the time when SO_2 emissions peaked around 1980. This process has been parameterized and implemented in the EMEP model, but there is still too little experimental data to thoroughly validate the magnitude of this effect. In total, however, the EMEP model represents the downward trend in sulfur over Europe for the last decades fairly well. Evaluations of the EMEP model using all present-day EMEP measurement data are available elsewhere [e.g., *Fagerli et al.*, 2003; *Simpson et al.*, 2006; *Jonson et al.*, 2006; H. Fagerli et al., manuscript in preparation, 2007].

[31] In the work by *Fagerli et al.* [2007], modeled sulfate concentrations were compared to sulfate concentrations derived from long-term ice core records at Col du Dome (CDD, French Alps) and Colle Gnifetti (CG, Swiss Alps). The modeled sulfate trend and the inverted summer ice core record were found to be in excellent agreement for CDD with a weak increase between 1920 and 1950, a steep increase toward the maximum around 1980, and then a significant decrease over the last two decades. The source apportionment performed in the paper showed that the largest contributors to the depositions of sulfate both at CDD and CG were Spain, France, Germany and Italy. At CG, the simulations and ice record were found to be in reasonable agreement from 1920 to 1970. After 1970, the

CG ice cores show a rather sudden decrease of concentrations, which was not found in the simulations, but this is possibly due to an artifact in the ice core record. In summary, the agreement between model simulations and the ice core records was rather encouraging and indicated that the historical emission inventories (at least for Spain, France, Germany and Italy) used for the study were reasonable.

3.3. Radiation Transfer Model

[32] The constructed trend of sulfate aerosol burden presented above was used as input for the Off-line Radiation Transfer Model (ORTM). A detailed model description is given by *Langmann et al.* [1998]. ORTM determines the direct and indirect shortwave forcing of sulfate aerosol based on the variable sulfate mass distribution and meteorological input data. The delta-Eddington approximation includes single as well as multiple scattering. Only the shortwave part of the solar spectrum, 0.2–5 μm , subdivided into 18 wavelength intervals, is considered, because sulfate aerosol has a negligible radiative effect in the infrared. Optical properties of dry sulfate aerosol are determined from Mie theory calculations, assuming a zero-order logarithmic size distribution $n(r)$ [*Lenoble and Brogniez*, 1984], with a geometric mean particle radius r_{gN} of 0.05 μm and a geometric standard deviation σ_g of 1.8. The particle density was set to 1.6 g cm^{-3} , which is equivalent to a mean particle composition of 75% H_2SO_4 and 25% H_2O . The modification of aerosol specific extinction due to the relative humidity of the ambient air (RH) is considered using a simple approximation adapted from the data given by *Nemesure et al.* [1995]. For relative humidity (RH) below 80%, the specific extinction is enhanced by a factor of $\text{RH} \cdot 0.04$, assuming a minimum relative humidity of 25%. For relative humidity exceeding 80%, the specific extinction increases exponentially with RH. The factor 9.9 is reached

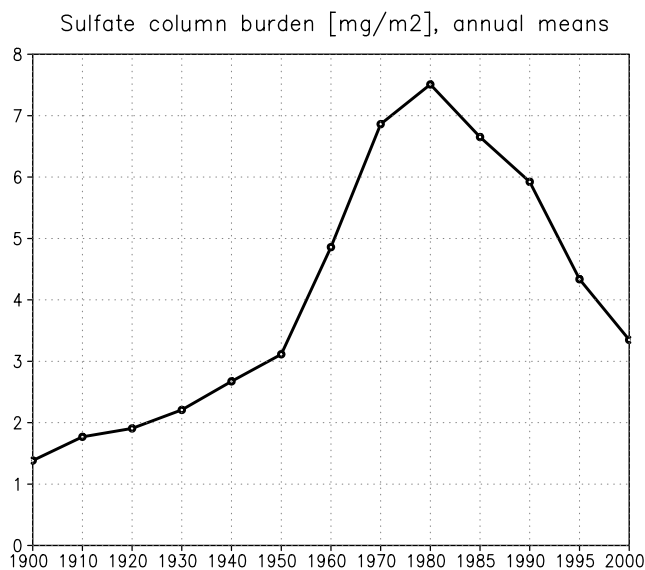


Figure 4. Total atmospheric sulfate load ($\text{mg}(\text{m}^{-2})$) over Europe, annual means, 1900–2000.

for $\text{RH} = 100\%$. The single scattering albedo and the asymmetry factor are assumed to be independent of RH . This approach might result in a small overestimation of the shortwave radiative forcing of sulfate aerosol, because with increasing relative humidity forward scattering is increased and backscattering in space direction reduced (asymmetry factor increased).

[33] For this study, we have calculated the shortwave direct radiative forcing of the total sulfate aerosol (natural and anthropogenic), for all sky conditions at the top of the atmosphere (TOA). The meteorological parameters which have important impacts on the direct aerosol forcing, the cloud cover and the relative humidity, can be obtained from *Benedictow* [2005], the surface albedo is shown in Figure 3.

4. Historical Trends

4.1. European Atmospheric Load of Sulfate Aerosol

[34] The atmospheric load of sulfate over Europe increased from the 1900s to the 1980s because of expanding industrialization since the early 20th century and uncontrolled emissions of SO_x (sulfur dioxide and sulfate). The sulfate load more than doubled during the first half of the century from $1.4 \text{ mg}(\text{m}^{-2})$ sulfate in the 1900s to $3.1 \text{ mg}(\text{m}^{-2})$ in the 1950s, with a very rapid growth in the following 20 years, when it again more than doubled reaching $6.7 \text{ mg}(\text{m}^{-2})$ in the 1970s (Figure 4).

[35] The maximum annual mean of the total column burden was reached in the 1980s, with $7.5 \text{ mg}(\text{m}^{-2})$ sulfate, more than 5 times higher than at the beginning of the century. The awareness of the health risks and environmental impacts of the atmospheric sulfate aerosol pollution resulted in emission control in western Europe in the 1980s, leading to a constant and significant reduction of anthropogenic SO_x emissions [*Mylona*, 1996]. Eastern Europe followed suit in the 1990s. From the 1980s until the present, the sulfate load has been decreasing. The mean

load in the year 2000 was $3.3 \text{ mg}(\text{m}^{-2})$, slightly higher than in the 1950s.

[36] The modeled amount of oxidants, ozone (O_3), hydrogen peroxide (H_2O_2) and hydroxy radicals (OH), have changed considerably during the century. O_3 and H_2O_2 have increased by a factor of 2–3. OH has increased by a factor of 2.5 on average in the boundary layer (while it also decreased over some areas like UK, Belgium, and some marine areas affected by ship traffic). Few other studies on the historical trends in oxidants have been published [*Lelieveld et al.*, 2002; *Wang and Jacob*, 1998; *Berntsen et al.*, 1997]. The increase in OH since preindustrial times based on our model calculations is clearly larger than in, e.g., the work by *Wang and Jacob* [1998] who found an increase of 40–60% over land in the northern latitudes. Sensitivity runs show that the reason for the increase in OH is the increase in the NO_x/CO emission ratio from the 1900s. In this study, we have used NO_x and CO emission inventories from different sources. In a test run, the NO_x emissions for 1900 were scaled to the same level as the ones used by *van Aardenne et al.* [2001] (the same source as used for our CO emissions). In this run, modeled OH concentration for 1900 was considerably higher (resulting in an increase of OH from the 1900s to the 1980s of about 60% over continental Europe, but still with large regional differences). The effect on sulfate concentration for the 1900s is around 5–10%, but larger (15–20%) for the area around Sicily, where the oxidation of high volcanic emissions is limited by oxidants. In the beginning of the century, SO_x emissions were low, and the level of oxidants was generally not a limitation for the oxidation to sulfate. The availability of oxidants becomes critical for the period when SO_x emissions reached their maximum (1970s/1980s). Our CO and NO_x emissions for this period stem from the same source (Table 2), hence the ratio is attributed to a lower uncertainty. We therefore conclude that the use of CO and NO_x emissions from different sources is a minor source of uncertainty in our model study for sulfate.

4.2. European Direct Radiative Forcing

[37] The temporal pattern of the direct aerosol forcing directly reflects the pattern of sulfate aerosol burden. The forcing is negative, because sulfate aerosol particles scatter incoming shortwave radiation. The annual negative maximum of the forcing is seen in July, while the minimum is found in December (Figure 5). During winter months, solar radiation is very weak in high latitudes and the aerosol forcing is almost negligible. Like the atmospheric sulfate load, the direct radiative forcing increased from 1900 to 1980, it more than doubled between the 1950s and the 1970s. After 1980 the direct forcing steadily decreased. The monthly mean of the sulfate direct shortwave forcing over Europe was -0.08 W m^{-2} in December 1900 and -0.4 W m^{-2} in December 1980, from when it constantly decreased to -0.2 W m^{-2} in December 2000, again comparable to December 1950. During summer, enhanced solar radiation results in a stronger forcing. The forcing is 5 to 7 times stronger in July than in December. The historical trend is much more pronounced in summer: from -0.6 W m^{-2} in July 1900 the forcing increased to -2.7 W m^{-2} in July 1980, and then it steadily decreased. In July 2000 the direct

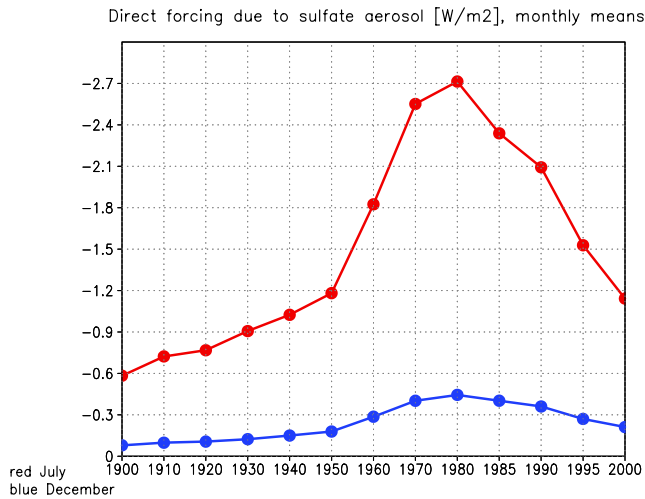


Figure 5. Direct shortwave radiative forcing due to sulfate aerosol (W m^{-2}) over Europe, 1900–2000, monthly means. Blue indicates December, and red indicates July.

sulfate forcing was reduced to -1.1 W m^{-2} , which is again comparable to the value in July 1950.

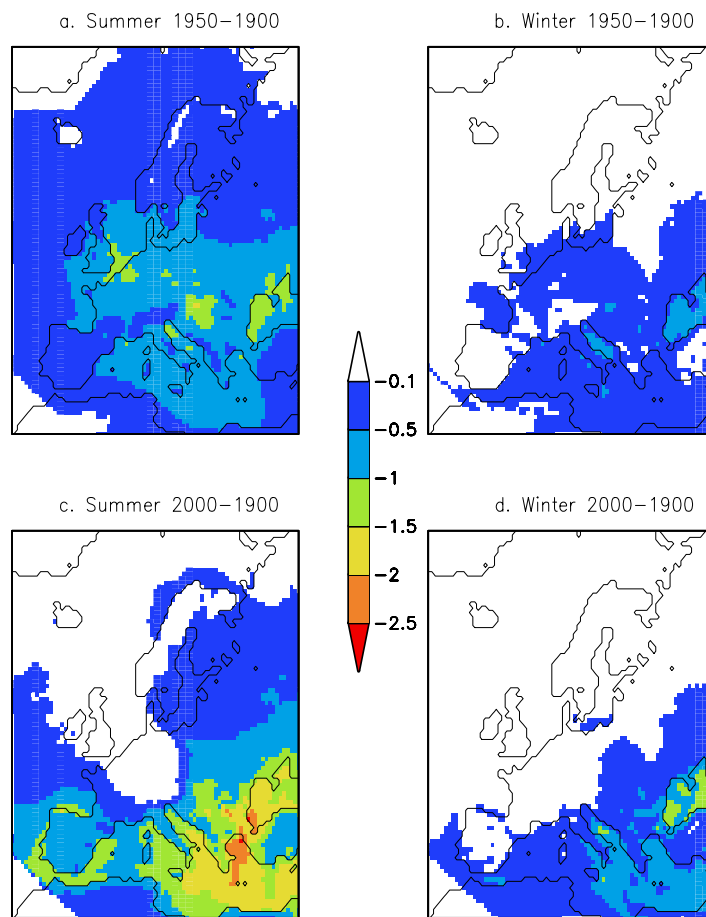
5. Regional Patterns of the Historical Trend

[38] In order to consider the regional aspects of the historical trend of the direct shortwave forcing of sulfate aerosol, we have analyzed the spatial evolution. The spatial evolution of the atmospheric load and the radiative forcing is discussed separately for winter (December, January, February) and for summer (June, July, August). Figure 6 illustrates the influence of the anthropogenic sulfate, showing the change in direct radiative forcing from 1900 to 1950 and from 1900 to 2000 for winter and summer, assuming that natural sources dominated the sulfate burden in 1900.

5.1. Winter

[39] During winter, photochemical processes responsible for the production of secondary pollutants are limited because of weak solar radiation intensity.

[40] In winter 1900, the highest atmospheric load can be found over central and southeastern Europe, mainly over Germany, with a seasonal mean over Germany of $5 \text{ mg(m}^{-2})$



Direct short wave radiative forcing of anthropogenic sulfate [W/m^2]

Figure 6. Change of the direct radiative sulfate aerosol forcing. (a) From 1900 to 1950, summer mean; (b) from 1900 to 1950, winter mean; (c) from 1900 to 2000, summer mean; and (d) from 1900 to 2000, winter mean.

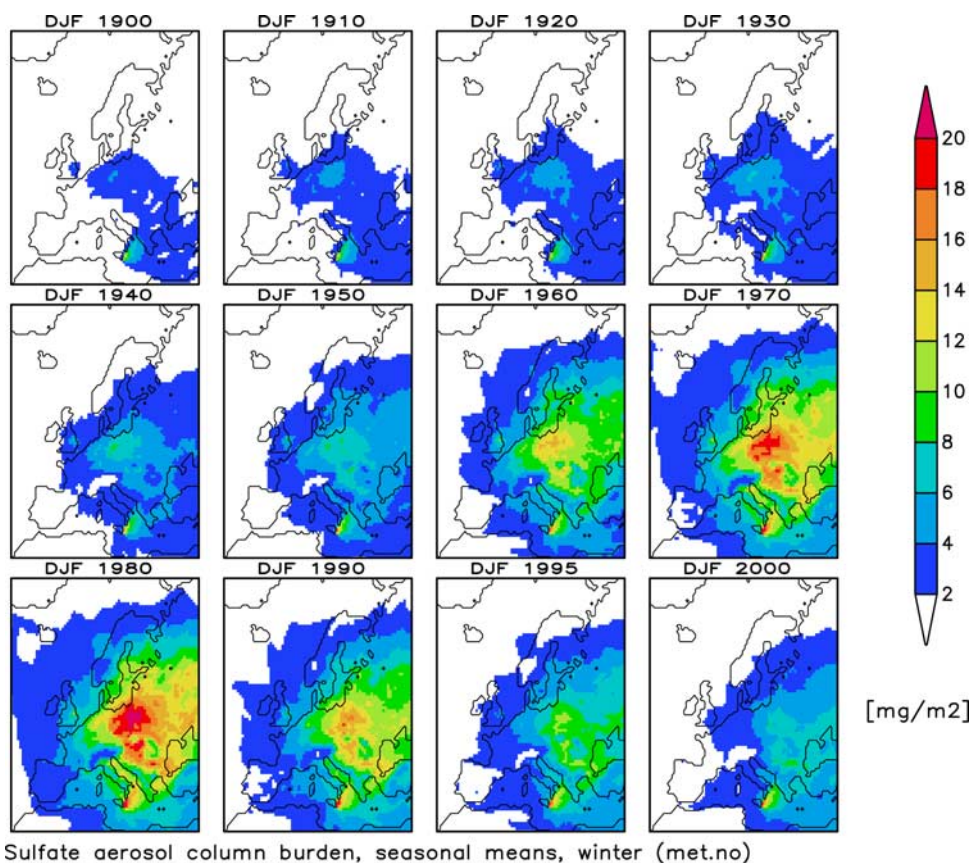


Figure 7. Wintertime horizontal distribution of the total atmospheric sulfate load ($\text{mg}(\text{m}^{-2})$), seasonal means, 1900–2000.

sulfate, compared with the mean European value of $1 \text{ mg}(\text{m}^{-2})$ (Figure 7). A second maximum is located over the Mediterranean Sea, which originates from the volcanic degassing of Mount Etna, a point source producing about $13 \text{ mg}(\text{m}^{-2})$ sulfate burden close to the source. The central European pollution maximum intensified and expanded northward, eastward and southward during the century. In the 1950s the maximum reached $7 \text{ mg}(\text{m}^{-2})$, in the 1980s over $18 \text{ mg}(\text{m}^{-2})$. The wintertime pollution over eastern Europe is mainly due to domestic heating with brown coal. The Mediterranean plume increased after the 1960s because of additional anthropogenic sources, including industry and traffic. During the 1980s, the European continent excluding northern Scandinavia and the Iberian Peninsula was very polluted during winter, with sulfate aerosol loads exceeding $14 \text{ mg}(\text{m}^{-2})$ over most of the continent. The pollution maximum was shifted from Germany toward the east: Poland, western Soviet Union, Czechoslovakia, Yugoslavia, Hungary and Bulgaria. From the 1980s until the year 2000, the sulfate pollution was significantly reduced, with mean values of $3 \text{ mg}(\text{m}^{-2})$ over western Europe, and $7 \text{ mg}(\text{m}^{-2})$ over eastern and southern Europe and the eastern Mediterranean Sea. The mean European atmospheric load in winter 2000 is close to that of the 1950s, but the regional distribution is different. In winter of the 1950s the pollution maximum was located over Germany and Poland, whereas it was shifted south eastward toward the Black Sea during winter 2000.

[41] The spatial evolution of the wintertime direct sulfate forcing does not follow that of the atmospheric load (Figure 8). Because of weak radiation in the higher latitudes, the forcing is almost negligible in these regions. The forcing is concentrated in the lower latitudes, over areas with high atmospheric aerosol load, predominantly over the Black Sea and the Mediterranean Sea. The scattering effect of aerosols is increased over surfaces with low surface albedo such as water. From seasonal mean of -0.3 W m^{-2} over these areas in 1900, it reached -1.8 W m^{-2} over the Mediterranean Sea and -2.5 W m^{-2} over the Black Sea in 1980. The mean December forcing in 2000 is slightly higher than in the 1950s, with a similar spatial pattern: It is concentrated in the lower latitudes with the strongest forcing observed over the Mediterranean Sea and the Black Sea. While some forcing is detectable over central Europe in winter in the 1950s, it is negligible in winter 2000. This is clearly due to reduced atmospheric load over western and central Europe in winter 2000 compared to the 1950s.

5.2. Summer

[42] During the summertime, photochemical processes responsible for the production of sulfate aerosol are enhanced because of strong solar radiation. In southern Europe, wet deposition is strongly limited by summertime aridity. These meteorological conditions cause pollution enhancement. Only in eastern Europe the summertime

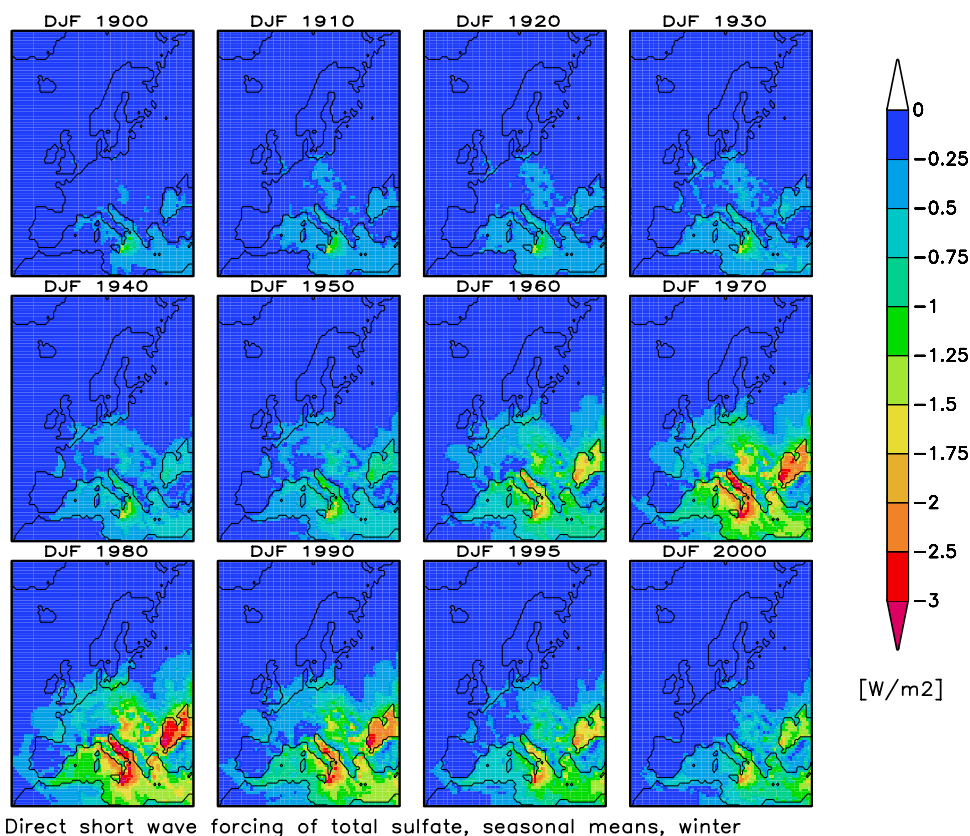


Figure 8. Wintertime direct radiative forcing due to sulfate aerosol (W m^{-2}), seasonal means, 1900–2000.

pollution is reduced, because there domestic heating is one of the main pollution sources.

[43] The spatial evolution of direct radiative forcing due to sulfate aerosol in summer is very similar to the spatial evolution of the sulfate atmospheric load (Figures 9 and 10). In summer 1900, two pollution maxima can be seen: one over northwestern Europe (Great Britain, Netherlands, Belgium, western Germany) and the North Sea and a second over the Mediterranean Sea. The northwestern plume intensified until the 1980s: The seasonal summertime mean increased from $6 \text{ mg(m}^{-2}\text{)}$ in 1900s to over $24 \text{ mg(m}^{-2}\text{)}$ in the 1980s; it expanded south eastward toward the Black Sea (Figure 9). The Black Sea was relatively unpolluted during summer in the first half of the century. The pollution enhanced rapidly from $3 \text{ mg(m}^{-2}\text{)}$ in the 1950s to $12 \text{ mg(m}^{-2}\text{)}$ in the 1980s. The Mediterranean pollution maximum expanded from the 1950s through the 1980s when the summertime mean value exceeded $22 \text{ mg(m}^{-2}\text{)}$. What we see now as a Sicilian plume is no longer dominated by the volcanic emissions from Mount Etna. The local anthropogenic emissions in Sicily peaked in the 1980s to become an important contributor to the plume. From the 1980s the summertime pollution over Europe began to decrease: The decrease of the Mediterranean plume is much slower than that of the plume over northwestern Europe. From 1995 on, the Mediterranean Sea is the most polluted area in summertime Europe. The main contributor to the summertime Mediterranean pollution nowadays are ship emissions. 54% of the total sulfate aerosol column burden

over the Mediterranean in summer originates from ship emissions, contributing more than 50% of the direct radiative forcing [Marmar and Langmann, 2005].

[44] The spatial pattern of the aerosol burden in summer is very well reflected in the spatial evolution of the direct sulfate aerosol forcing (Figure 7). The forcing over the North Sea and western Europe enhanced steadily since the 1900s. Until the 1970s, the forcing was strongest over northwestern Europe with mean summer values exceeding -6.5 W m^{-2} . Afterward the forcing over the Mediterranean became dominant with values of up to -6 W m^{-2} . The forcing over the Black Sea was very high from 1970 until 1990. By the year 2000, the summer mean direct forcing over northwestern Europe was -1.5 to -2 W m^{-2} , while over the central Mediterranean Sea, particularly over the Aegean Sea, it reached -3.5 W m^{-2} . While the mean forcing in July 2000 has a similar value as July 1950, its maxima shifted from the North Sea to the Mediterranean Sea. In July 2000, the forcing over the North Sea was -1.5 W m^{-2} , compared to -5 W m^{-2} in the 1950s.

5.3. Forcing Efficiency

[45] The concept of “forcing efficiency,” defined as the ratio between the direct radiative forcing and the column burden was introduced by Boucher and Anderson [1995]. We found the modern European mean forcing efficiency to be $-230 \text{ W(g sulfate)}^{-1}$. It is well within the range of mean global sulfate forcing efficiencies from different simulations and methods of -130 to $-370 \text{ W(g sulfate)}^{-1}$ [Seinfeld, 2002]. In the AeroCom experiment, sulfate forcing efficien-

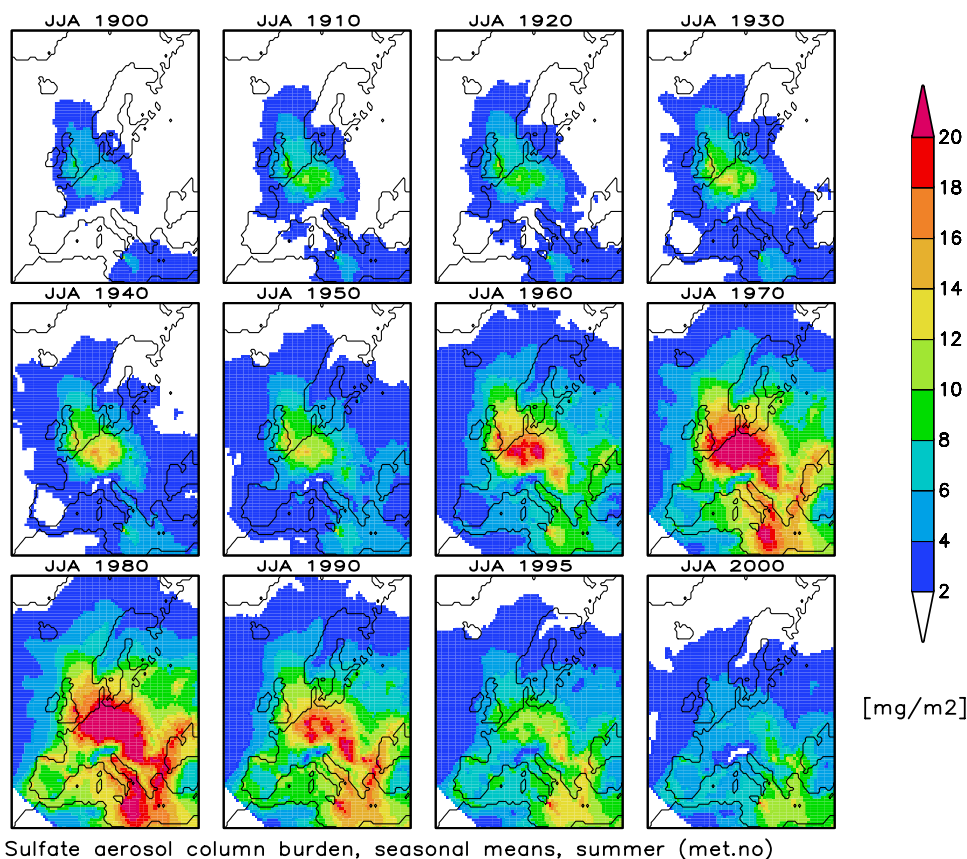


Figure 9. Summertime horizontal distribution of the total atmospheric sulfate load ($\text{mg}(\text{m}^{-2})$), seasonal means, 1900–2000.

cies have been predicted by different global models using prescribed SO_x emissions, the global annual means ranging from -134 to $-233 \text{ W}(\text{g sulfate})^{-1}$ [Schulz *et al.*, 2006]. According to our findings, the forcing efficiency depends strongly on the season, the latitude and the surface albedo. In our model, the mean forcing efficiency is $-78 \text{ W}(\text{g sulfate})^{-1}$ in December and $-335 \text{ W}(\text{g sulfate})^{-1}$ in July, averaged from 1900–2000. These results qualitatively agree with the study of Boucher and Anderson [1995], with forcing efficiency of $-62 \text{ W}(\text{g sulfate})^{-1}$ for central Europe in January, and of $-193 \text{ W}(\text{g sulfate})^{-1}$ for July. Boucher and Anderson [1995] also found a pronounced negative correlation between the forcing efficiency and the cloud cover fraction. Climatological dependencies of the forcing efficiency could not be analyzed in this study, because the same meteorological year was used for all simulations. Additionally, the forcing efficiency depends the influence of the relative humidity on the aerosol optical properties. The forcing efficiency is not a temporally constant value. In December, the forcing efficiency in 2000 with $-83 \text{ W}(\text{g sulfate})^{-1}$ is higher than in the 1900s with $-76 \text{ W}(\text{g sulfate})^{-1}$ (Figure 11), because of the shift of sulfate aerosol burden maxima from higher to lower latitudes. The December forcing efficiency has a minimum of $-69 \text{ W}(\text{g sulfate})^{-1}$ in the 1960s. This is because the aerosol burden in the higher latitudes, which results in no or very low forcing during winter, initially increased faster than in the low latitudes. This trend was reversed since the 1960s. In July, the forcing efficiency is reduced from -363 in the 1900s to

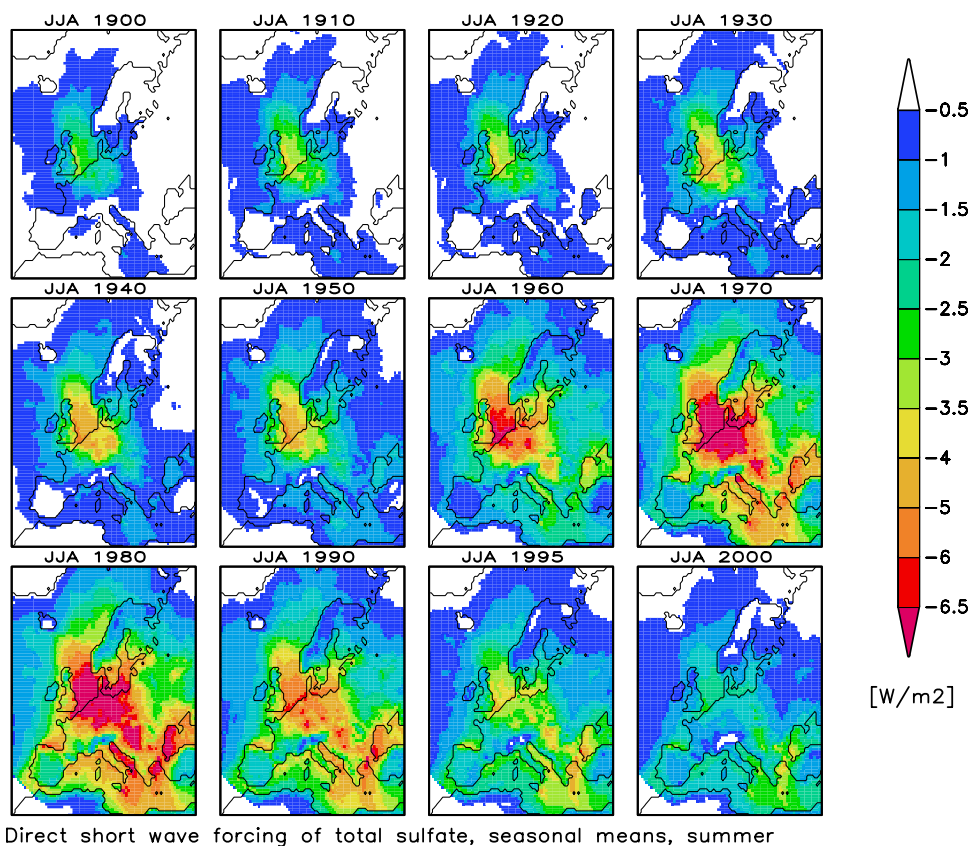
$-305 \text{ W}(\text{g sulfate})^{-1}$ in 2000. This reduction is caused by the shift of the pollution maxima from north to south. During the summertime, there are longer hours of daily solar radiation in the north, resulting in higher forcing efficiency. The annual mean forcing efficiency also slightly reduced from -246 to $-230 \text{ W}(\text{g sulfate})^{-1}$. The global mean historical forcing efficiency remained nearly constant in the work of Boucher and Pham [2002]. Probably, increased and decreased forcing tendencies compensate on the global mean.

5.4. Direct Forcing at Selected Areas

[46] Five different areas were selected to further illustrate different regional patterns of the direct radiative sulfate forcing: the English Channel, the Black Sea, Denmark, the Island of Sicily and Sonnblick, an Alpine mountain site.

[47] Surface albedo is lower over water than over land, resulting in stronger forcing efficiency over maritime areas. The present day forcing efficiency over the Black Sea is $-385 \text{ W}(\text{g sulfate})^{-1}$ in July and $-190 \text{ W}(\text{g sulfate})^{-1}$ in December, and over the English Channel $-440 \text{ W}(\text{g sulfate})^{-1}$ in July (Table 3).

[48] These values are much higher than the European mean. In winter, the English Channel receives less solar radiation than the Black Sea, resulting in lower efficiency of $-70 \text{ W}(\text{g sulfate})^{-1}$ in December. Seasonality is also very pronounced over Denmark, our most northern area, with strong forcing efficiency in July and weak in December.



Direct short wave forcing of total sulfate, seasonal means, summer

Figure 10. Summertime direct radiative forcing due to sulfate aerosol (W m^{-2}), seasonal means, 1900–2000.

[49] On the contrary, the forcing efficiency at the Island of Sicily, our most southern area, shows no pronounced seasonality ($-190 \text{ W}(\text{g sulfate})^{-1}$ in July and $-157 \text{ W}(\text{g sulfate})^{-1}$ in December). Relative to the European mean, the efficiency is low in summer, because of shorter days in the south than in the north, and high in winter, for the opposite reason. Over Sonnblick (3106 m asl) in December, the surface albedo of the snow covered mountains is high and the additional scattering by aerosols does not result in a significant forcing efficiency. The atmospheric burden of sulfate aerosol is the lowest at this elevated site; this site represents free tropospheric conditions. The resolution of the model results in a lower grid box elevation (2060 m asl), hence the simulated influence of the sulfate aerosol pollution from northwestern Europe is stronger than we would expect at this remote elevated region. The July forcing has a maximum in the 1985, and by the year 2000, this forcing is smaller than at the beginning of the century (Figure 12). The forcing over the English Channel, which is located close to strong sulfur emission sources, reaches -5.2 W m^{-2} in the 1970s, by the year 2000 it also reduced close to the values of the 1900s. Similar trend can be found over Denmark. Comparing the trends over Sonnblick, Denmark and the English Channel to that over the Black Sea, reveals the shift of the forcing maxima from northwestern to southeastern Europe (section 5.3). The modern forcing over the Black Sea with -2.5 W m^{-2} in July is 10 times stronger than at the beginning of the last century; the modern forcing in December is 5 times stronger than in the 1900s. The modern

forcing over the Black Sea is similar to that in the 1960s, its reduction in the past 30 years less efficient than that of the European mean forcing. The forcing over Sicily shows the

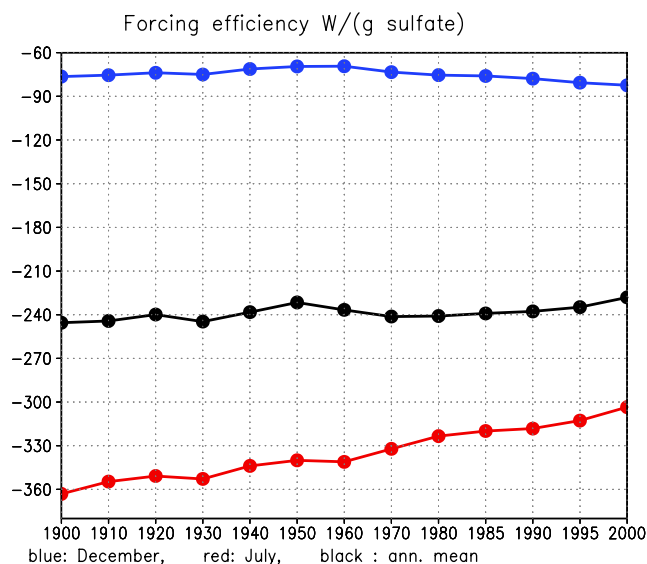


Figure 11. Historical trend in the forcing efficiency of sulfate aerosol over Europe ($\text{W}(\text{g sulfate})^{-1}$), 1900–2000. Black indicates yearly mean, blue indicates monthly mean December, and red indicates monthly mean July.

Table 3. Forcing Efficiencies for the Selected Areas in July and in December 2000

| Area | Forcing Efficiency, $W(g \text{ sulfate})^{-1}$ | |
|------------------|---|---------------|
| | July 2000 | December 2000 |
| English Channel | -440 | -70 |
| Black Sea | -385 | -190 |
| Denmark | -360 | -72 |
| Sonnblick | -212 | -33 |
| Island of Sicily | -190 | -157 |
| European mean | -338 | -78 |

same trend as over the Black Sea, but the absolute values are much lower.

5.5. Contribution of Ship Emissions

[50] Unlike the land based sulfur emissions, which experienced significant reduction due to environmental policies since the 1980s, ship emissions keep increasing at an annual rate of $\sim 2.5\%$ (Endresen *et al.* [2003] and Table 1). The sensitivity study without ship emissions has showed, that their relative contribution to the direct radiative forcing over Europe has increased from 2% in the 1950s to 10% in 2000 (Figure 13). This contribution is especially relevant over the western Mediterranean [Marmar and Langmann, 2005] and

southwest of the Gulf of Biskaya during summer, where it reaches 30% (Figure 14).

6. Summary and Conclusions

[51] We have provided an estimate of the historical evolution of the direct radiative forcing of sulfate aerosol over Europe, emphasizing on regional characteristics. The mean direct forcing has increased since the 1900s reaching its peak in the 1980s and then returning in present times to approximately the values of the 1950s. Despite the different distribution of the atmospheric load in December 1950 and 2000, the winter forcing distribution remains very similar, with maxima over the Black and the Mediterranean Seas. We found pronounced shift of the summer forcing maxima from northwestern to southeastern Europe. We can clearly observe that emission reductions, introduced in the 1980s, have led to a significant reduction in the atmospheric load and the direct forcing over Europe. The regional direct aerosol forcing depends not only on the sulfate load, but also on the latitude, the season, the cloud cover and the surface albedo. Ship emissions are found to increasingly contribute to sulfate aerosol burden and direct forcing, since their trend from the 1950s to 2000 is reverse to that of the land based emission sources.

[52] An uncertainty that needs to be carefully considered is the assumption of the same meteorological year [1997]

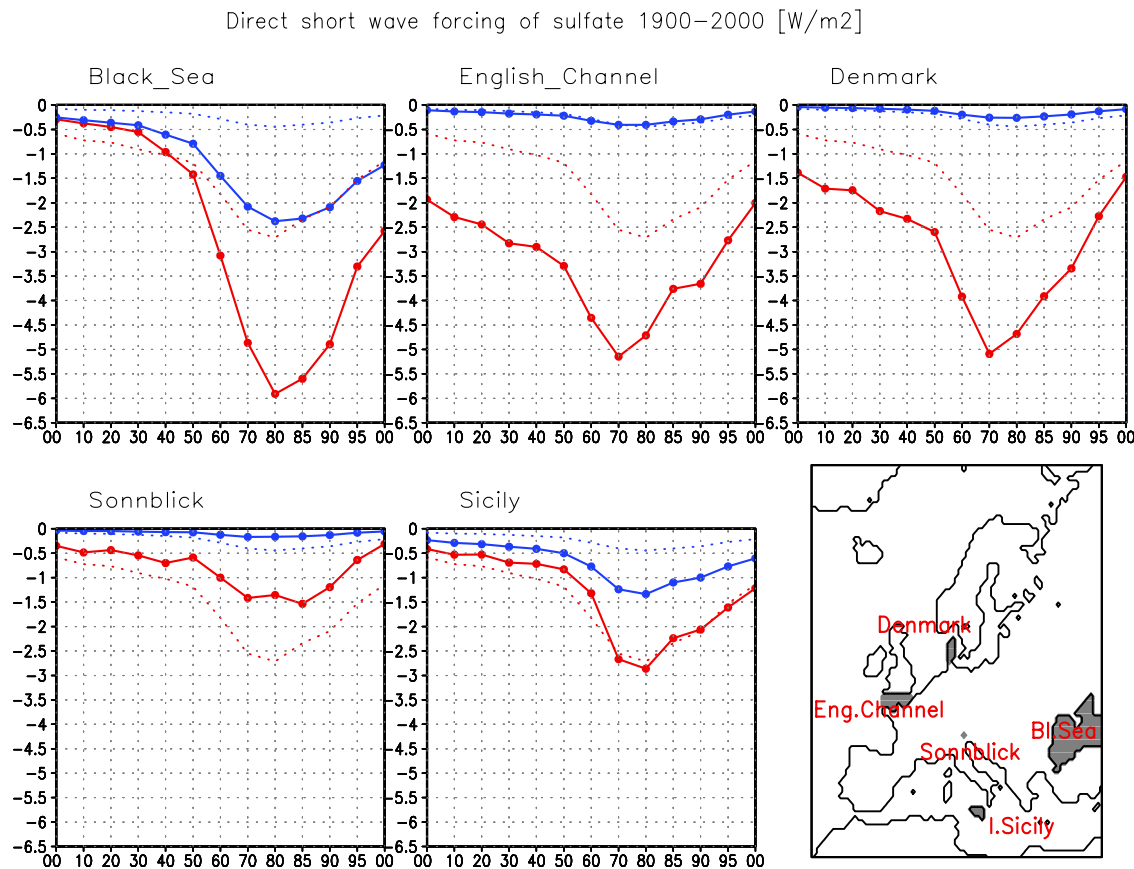


Figure 12. Historical trend of the direct sulfate forcing at the selected areas for July (red) and December (blue). Dotted lines show the trend of the European mean forcing in July (red) and December (blue). Also shown is a map of the selected areas.

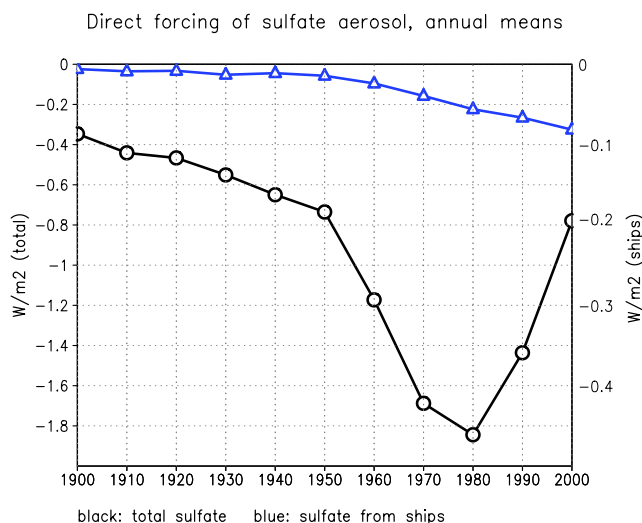


Figure 13. Historical trend of the direct radiative forcing due to total sulfate (black) and due to sulfate from ship emissions (blue).

for all simulations. Meteorological conditions play a very important role in aerosol production, transport and deposition [Marmier and Langmann, 2007] and can result in interannual concentration variabilities of up to 30% [Putaud et al., 2004]. Furthermore, meteorology has an important impact on the forcing itself, via changes in surface albedo due to snow cover, changes in cloud cover and relative humidity of the ambient air. The aerosol forcing thus depends not only on the emissions strength, but also on interannual meteorological variability, which was not considered. For example, negative precipitation trends over areas with increasing emissions (central Europe and Mediterranean) as indicated in IPCC [2001b] might have

resulted in higher atmospheric loads than modeled, while positive precipitation trends over Scandinavia and northern Russia could have the opposite effect. The trend in cloud cover [Warren et al., 2007] follows that in precipitation, enhancing the forcing efficiency in areas with less clouds and vice versa. The forcing efficiency could have been further enhanced by the reduction of snow cover and glaciers over the past century [Haeberli, 2003; Dyurgerov, 1999]. All these trends are of regional character and their impacts interact with each other, we can only speculate on their impact on the historical sulfate aerosol forcing.

[53] Additional uncertainty is caused by the treatment of RH in the radiation model. In ORTM, size-dependent Mie calculations have been applied for dry aerosol particle size. The specific extinction obtained for a dry particle was then multiplied with a RH-dependent growth factor. This approach may underestimate the direct radiative forcing for high relative humidities.

[54] With black carbon to be included in future work, we expect different historical evolution of the aerosol forcing distribution and strength. Black carbon is highly absorbing and so its radiative forcing is positive in sign. Thus the forcings might partially offset each other. Historical changes of the aerosol single scattering albedo as an indicator of aerosol direct forcing due to changes in the aerosol composition for different regions are estimated by Novakov et al. [2003]. The nonlinearity of the aerosol burden response associated with emission changes as suggested by Stier [2005] can additionally affect the historical aerosol burden and the corresponding direct forcing when black carbon is included. On seasonal and regional scales the sign of the total forcing might vary substantially. Historical gridded emission inventory of carbonaceous aerosols for Europe needs to be established in order to complete the model simulations of the direct aerosol forcing evolution.

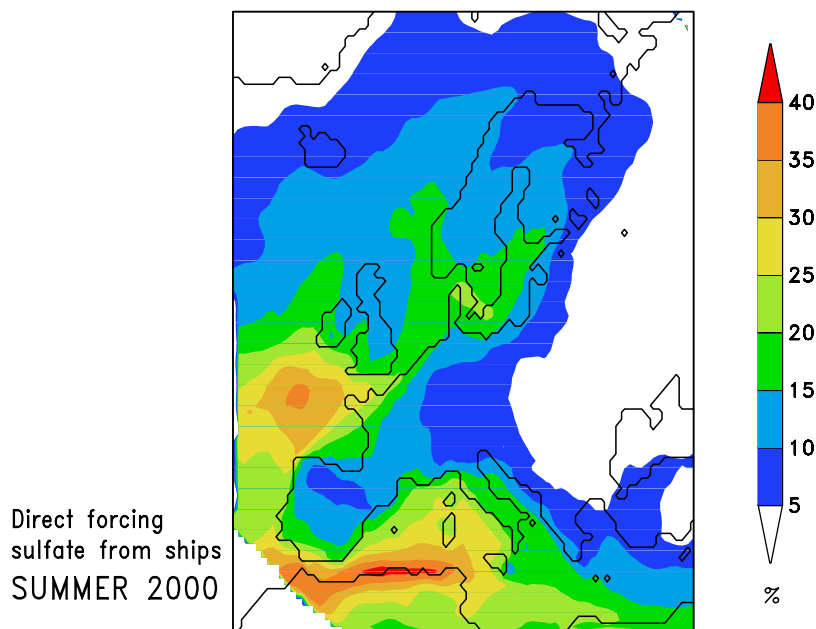


Figure 14. Seasonal mean of the relative contribution of sulfate from ships to the direct radiative forcing (%), summer 2000.

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