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Transboundary Acidification, Eutrophication and Ground Level Ozone in Europe in 2003

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Executive Summary

2003 has been characterized as a year with extreme weather conditions, due to the very warm and dry summer in Central Europe. The meteorological situation in summer 2003 was characterised by the extension of the semi-arid conditions from Mediterranean regions and the presence of persistent anti-cyclonic systems over central Europe.

This report, prepared in time for presentation at the 28th session of the Steering Body to EMEP, analyses the effect of such extreme meteorological conditions in the air concentrations of ozone and particulate matter and depositions of eutrophying and acidifying compounds, and analyses further how meteorological variability affects transboundary air pollution.

Was 2003 an extreme meteorological year?

Both measurements and model calculations have demonstrated that the summer of 2003 was indeed extreme in Central Europe. Some regions (France, northern Spain, Switzerland and northern Italy) were more than 4 degrees warmer in 2003 than in 2002. At the same time, the amount of precipitation was much lower in most of these regions, except parts of France. As a result, summer concentrations of ozone and secondary inorganic particles (SIA) increased substantially. For some regions, like for instance in Switzerland, summer concentrations of SIA were up to 60-80% higher than the previous year.

In yearly average, the meteorological conditions of 2003 give variations in the air quality indicators that are larger than for any of the previous years, but still comparable to other years in certain parts of Europe. These extreme 2003 meteorological conditions result in SOMO35 values about 10% higher that average, air concentrations of particulate matter also higher (about 35-50%) and depositions generally 20% lower. The generally lower deposition of sulphur and nitrogen results in considerably lower exceedances of critical loads for eutrophication in Europe in 2003.

Still, the characterisation of 2003 with respect to future climate indicates that we can expect an increase in the inter-annual variability of European climate conditions in response to greenhouse gas forcing. This means an increase in the probability of occurrence of extreme years like 2003 relative to mean climatic conditions. Even with a small mean temperature increase, climate model simulations suggest that, toward the end of this century, about every second summer could be as warm and dry in central Europe as 2003.

Did the EMEP model manage to reproduce the pollution levels in 2003?

In general, the EMEP Unified model performance is similar for 2003 than for previous years. Correlation coefficients and biases are similar to previous verification results for all components and subject to the same limitations that have already been reported.

For ozone, for instance, the general findings are that the model performs better for 2002 than for the year 2003 in terms of mean ozone, AOT40 and SOMO35, but that correlation coefficients were higher in 2003, often with $r > 0.8$. There are many possible reasons for the under-predictions found in the summer of 2003, including problems in specifying boundary conditions (BCs), both from tropospheric and stratospheric ozone, and the lack of response to soil moisture effects in the standard EMEP model. Soil moisture deficits can induce large reductions in the deposition velocity of ozone, and this should lead to higher ozone concentrations. Further work to elucidate the influence of these different processes will also contribute to determine the validity of the model and its predictions for future conditions.

How should we deal with meteorological variability under the revision of the Gothenburg Protocol?

It is proposed to use five meteorological years: 1996, 1997, 1998, 2000 and 2003 to support to review of the National Emission Ceilings Directive and the Gothenburg Protocol. These years have been selected in terms of their availability as input for transport model calculations, their climatological representativity over the last 30 years and with regard to feasible meteorological situations predicted for 2020-2030.

The first four years have been proposed because their average is closer to the European average meteorological conditions than any individual year and that is the case for all air quality indicators. So, for instance, $PM_{2.5}$ air concentrations averaged for the proposed four years (1996-1998, 2000) are generally over Europe within 10% difference from the 32-years average of the meteorological conditions from 1970 to 2002. For summer ozone, differences are below 5%. The year 2003 is proposed in addition, to represent a situation with larger variability. Current understanding of future climate situations indicates that these meteorological conditions would be more probable in the next 20-30 years and are expected to be within the statistical normal variability by 2070-2100.

How does meteorological variability affect transboundary air pollution?

The total contribution of transboundary fluxes to air pollution concentrations and depositions is relatively robust with respect to inter-annual variability. For all the analysed meteorological years, in most countries, the transboundary contributions to SOMO35 can be as high as 75% , whereas the transboundary contributions to PPM_{coarse} concentrations are usually lowest, often below 40%.

Inter-annual meteorological variations are significant however for individual country contributions. The individual contributions vary much more than the pollutant levels itself, with variabilities as high as 40%. This affects the optimisation results from integrated assessment modelling and, given the large inter-annual variations in individual contributions, it is recommended that this variability is taken into account by Integrated Assessment Modelling (IAM), either by considering average source receptor matrices or by performing the optimization process for a number of different meteorological years and considering the average results.

Could meteorological variability dominate over emission reductions in 2020?

The effect of meteorological variability in air pollution levels might mask the benefits of emission reductions projected for 2020 in large areas over Northern and Eastern Europe, both for ozone and for particulate matter. The projected emission reductions for 2020 yield substantial improvements of environmental quality in Europe. However, even with additional control measures, the targets for health and the environmental standards can not be met over large parts of Europe. In particular, ozone future levels will also depend on hemispheric background ozone. Changes in background levels can be caused by changes in emissions in other continents or in changes in circulation, an effect that that has not been considered in the present calculations and requires further evaluation efforts.

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Contents

1 Introduction **1**

1.1 Overview 1
1.2 Definitions, statistics used 2
1.3 Country Codes 5
1.4 Other Publications 5
References 8

2 Acidifying and eutrophying components, status in 2003 **9**

2.1 Comparison of observations and model results for 2003. Was 2003 a special year? 9
2.1.1 Summary of model performance for 2003 10
2.2 2003 versus 2002 18
2.2.1 Can the model tackle extreme conditions? 23
2.3 Deposition of sulphur and nitrogen in 2003 and exceedances of critical loads 25
References 30

3 Particulate Matter concentrations, status in 2003 **33**

3.1 Model results for PM 33
3.1.1 Concentration fields in 2003 33
3.2 European aerosol concentrations in 2003 compared to 2002 36
3.2.1 Effect of emission changes between 2002 and 2003 36
3.2.2 Effect of meteorological variability between 2003 and 2003 . 38
3.3 Evaluation of the model performance for 2003 42
3.3.1 Annual mean scatter plots 42
3.3.2 Monthly variation of aerosol concentrations 44

3.3.3 Model calculations of PM under the heat wave conditions in 2003 . . . 47
References . . . 48

4 Photo-oxidants, status in 2003 51
4.1 Introduction . . . 51
4.2 Time-series for ozone . . . 52
4.2.1 Nordic Sites . . . 55
4.2.2 Eastern European Sites . . . 55
4.2.3 Central and North-West European Sites . . . 55
4.2.4 Mediterranean Sites and Portugal . . . 63
4.3 Other statistics, O_3 . . . 64
4.4 Nitrogen Dioxide . . . 67
4.5 Discussion and Conclusions . . . 69
References . . . 69

5 Meteorological years available for use under the review of the National Emission Ceilings Directive (NECD) and the Gothenburg Protocol 73
5.1 Introduction . . . 73
5.2 Analysis of the meteorological variability of different pollutants over 9-years . . . 74
5.3 Climatological characterisation of 1996, 1997, 1998 and 2000 . . . 77
5.4 Inclusion of 2003 as extreme year? . . . 81
5.5 Consequences for future control strategies . . . 82
5.6 Conclusions and recommendations . . . 85
References . . . 87

6 Meteorological variability in source allocation: Transboundary contributions across Europe 89
6.1 Introduction . . . 89
6.2 Transboundary versus indigenous contributions . . . 91
6.3 Meteorological variability of SR calculations . . . 94
6.3.1 Analysis on country level . . . 97
6.3.2 Analysis on grid level . . . 101
6.3.3 The year 2003 . . . 103
6.4 SR results for the Appendix . . . 106
6.5 SR results for the country reports . . . 106
6.6 Conclusions and recommendations . . . 106
References . . . 107

7 Air quality indicators for CAFE scenarios 109
7.1 Introduction . . . 109
7.2 Description of the scenarios . . . 110

7.3 Status in 2000, CAFE-Baseline . . . 112
7.3.1 Ozone . . . 112
7.3.2 $PM_{2.5}$ . . . 113
7.3.3 Depositions of sulphur and nitrogen compounds . . . 114
7.4 Effects of emission projections for 2010 . . . 115
7.4.1 Ozone . . . 115
7.4.2 $PM_{2.5}$ . . . 115
7.4.3 Depositions of S and N compounds . . . 115
7.5 Effect of emission projections for 2020 . . . 115
7.5.1 Ozone . . . 116
7.5.2 $PM_{2.5}$ . . . 117
7.5.3 SIA speciation . . . 118
7.5.4 Depositions of sulphur and nitrogen . . . 118
7.6 Benefits of the emission reductions . . . 122
References . . . 125

8 Intercontinental air pollution transport 127
8.1 Major transport mechanisms . . . 128
8.1.1 ICT of natural emissions . . . 131
8.1.2 Influence of ICT on lower tropospheric concentration levels . 132
8.2 Studies on ICT within EMEP: Assessment of the impact of ICT on European surface pollution levels . . . 133
8.3 Hemispheric/global version of the Unified EMEP model . . . 134
8.3.1 Evaluation of new meteorological input data: ERA-40 data . . 135
8.3.2 Effect of the new meteorology on the dispersion of air pollutants 141
8.3.3 Towards a hemispheric model application . . . 145
References . . . 147

A National emissions and projections A:1

B Source-Receptor relationships: Tables for 2010 B:1

C Source-Receptor relationships: Tables for 2003 C:1

CHAPTER 1

Introduction

1.1 Overview

On the 17th May 2005, the Gothenburg Protocol entered into force and with this the process of review of the Protocol was initiated. The review of the Gothenburg Protocol coincides then in time with the review of the National Emission Ceilings Directive (NECD) and the two processes will share similar challenges. Among of these, is the challenge of how to deal with environmental protection in a world affected by climate variability. This implies not only analysing the synergies between control strategies for air pollutants and greenhouse gases but also understanding how dynamic and physical changes in the climate system are affected by air pollution and how these influence in turn the transport and composition of air pollution.

Triggered by the situation in 2003, characterized by extreme summer weather conditions in Central Europe,the analysis of meteorological variability has become a priority for the EMEP centers in the initial stages of the work for the revision of the Gothenburg Protocol.

The first three chapters in this report analyse the air pollution situation in 2003 for acidifying and eutrophying compounds, atmospheric particles and ozone, respectively. All three chapters compare the air pollution situation in 2003 with results in 2002 and some previous years, aiming at characterizing to what degree 2003 was indeed a extreme year and to what extent the EMEP model is able to reproduce such extreme situations.

The next chapter, Chapter 5, evaluates the meteorological conditions of 2003 in a broader context and analyses the effect of effect of meteorological variability in ozone, PM and sulphur and nitrogen deposition levels over the last 33 years. It proposes to take into account the increasing climatological variability by selecting 5 different me-

teorological years to be used under review of the NECD and the Gothenburg Protocol. The requirement for these five years is that they provide a reasonable representation of the expected meteorological conditions over the next two decades.

Based on the selected five meteorological years, Chapter 6 evaluates the expected effect of meteorological variability in transboundary source allocation. Although the total contribution of transboundary fluxes remains relatively constant, inter-annual meteorological variations are significant for individual country contributions. This affects the optimisation results from integrated assessment modelling and it is widely recognized that future scenario analysis requires to consider meteorological variability. The results have already been used by the CAFE programme and short presentation of the air quality levels projected for 2010 and 2020 according the CAFE scenarios is presented in Chapter 7.

The last chapter in the report deals with the concern that regional control strategies may be inefficient as background concentrations of air pollutants increase due to rising emissions in other parts of the world. The new Task Force on Hemispheric Transport of Air Pollution (TFHTAP) under EMEP will work to plan and conduct scientific work necessary to develop a fuller understanding of intercontinental transport of air pollution for consideration in the review of Protocols to the Convention and for that it will rely on the expertise of a broad scientific community. Chapter 8 introduces how the EMEP centers are prepared to contribute to the work of this Task Force.

All data included in this report will be available at the EMEP web site after its presentation at the 29th session of the EMEP Steering Body. Countries are encouraged to analyse the data and provide their own conclusions. Reactions and comments are both welcome and encouraged.

Description of the EMEP Unified model and its results, including source-receptor calculations, can be obtained from the EMEP web-site, http://www.emep.int.

1.2 Definitions, statistics used

For sulphur and nitrogen compounds, the basic units used throughout this report are μg (S or N)/m^3 for air concentrations and mg (S or N)/m^2 for depositions.

This report includes also concentrations of particulate matter (PM). The basic units throughout this report are μg/m^3 for PM concentrations and the following acronyms are used for different components to PM:

SIA - are secondary inorganic aerosols and are defined as the sum of sulphate (SO_4), nitrate (NO_3) and ammonium (NH_4). In the Unified EMEP model SIA is calculated as the sum: SIA= SO_4 + NO_3(fine) + NO_3(coarse) + NH_4

PPM - denotes primary particulate matter, originating directly from anthropogenic emissions. It is usually distinguished between fine primary particulate matter,

$PPM_{2.5}$, with dry aerosol diameters below 2.5 μm and coarse primary particulate matter, PPM_{co}, with dry aerosol diameters between 2.5μm and 10μm.

$PM_{2.5}$ - denotes fine particulate matter, defined as the integrated mass of aerosol with dry diameter up to 2.5 μm. In the Unified EMEP model $PM_{2.5}$ is calculated as the sum: $PM_{2.5}$ = SO_4 + NO_3(fine) + NH_4 + $PPM_{2.5}$

PMcoarse - denotes coarse particulate matter, defined as the integrated mass of aerosol with dry diameter between 2.5μm and 10μm. In the Unified EMEP model PMcoarse is calculated as the sum: PMcoarse = NO_3(coarse) + PPM_{co}

PM_{10} - denotes particulate matter, defined as the integrated mass of aerosol with dry diameter up to 10 μm. In the Unified EMEP model PM_{10} is calculated as the sum: PM_{10} = SO_4 + NO_3(fine) + NH_4 + $PPM_{2.5}$+NO_3(coarse) + PPM_{co}

For ozone, the basic units used throughout this report are ppb (1 ppb = 1 part per billion by volume) or ppm (1 ppm = 1000 ppb). At 20°C and 1013 mb pressure, 1 ppb ozone is equivalent to 2.00 μg m^{-3}.

A number of statistics have been used to describe the distribution of ozone within each grid square:

Mean of Daily Max. Ozone - First we evaluate the maximum modelled concentration for each day, then we take the 6-monthly mean of these values, over the 6-month period 1 April - 30 September.

SOMO35 - The Sum of Ozone Means Over 35 ppb is the indicator for health impact assessment recommended by WHO. It is defined as the yearly sum of the daily maximum of 8-hour running average over 35 ppb. For each day the maximum of the running 8-hours average for O_3 is selected and the values over 35 ppb are summed over the whole year.

If we let A_8^d denote the maximum 8-hourly average ozone on day d, during a year with Ny days (N_y = 365 or 366), then SOMO35 can be defined as:

$SOMO35 = \sum_{d=1}^{d=Ny} max\left(A_8^d - 35 \text{ ppb}, 0.0\right)$

where the max function ensures that only A_8^d values exceeding 35 ppb are included. The corresponding unit is ppb.days.

AOT40 - the accumulated amount of ozone over the threshold value of 40 ppb, i.e..

$$AOT40 = \int max(O_3 - 40 \text{ ppb}, 0.0)\, dt$$

where the max function ensures that only ozone values exceeding 40 ppb are included. The integral is taken over time, namely the relevant growing season for the vegetation concerned. The corresponding unit are ppb.hours (abbreviated to ppb.h). The usage and definitions of AOT40 have changed over the years though, and also differ between UNECE and the EU. Mills (2004) give the latest definitions for UNECE work, and describes carefully how AOT40 values are best estimated for local conditions (using information on real growing seasons for example), and specific types of vegetation. Further, since O_3 concentrations can have strong vertical gradients, it is important to specify the height of the O_3 concentrations used. In previous EMEP work we have made use of modelled O_3 from 1 m or 3 m height, the former being assumed close to the top of the vegetation, and the latter being closer to the height of O_3 observations. In the Mapping Manual (Mills 2004) there is an increased emphasis on estimating AOT40 using ozone levels at the top of the vegetation canopy.

Although the EMEP model now generates a number of AOT-related outputs, in order to allow great flexibility in later analysis, we will concentrate in this report on four "practical" definitions:

AOT40$^{\mathbf{3m}}$ - AOT40 calculated from O_3 concentrations at 3 m height. This AOT40 is close to that derived from measurements. (Technically, the 3 m is above the displacement height, and so close to the top of a forest canopy, but well above a crop canopy).

AOT40$^{\mathbf{3m}}_{f}$ - AOT40 calculated as above, but over April-September in analogy with previous calculations of AOT40f for forests.

AOT40$^{\mathbf{uc}}_{f}$ - AOT40 calculated for forests using estimates of O_3 at forest-top (uc: upper-canopy). This AOT40 is that defined for forests by Mills (2004), but using a default growing season of April-September.

AOT40$^{\mathbf{uc}}_{c}$ - AOT40 calculated for agricultural crops using estimates of O_3 at the top of the crop. This AOT40 is close to that defined for agricultural crops by Mills (2004), but using a default growing season of May-July, and a default crop-height of 1 m.

In all cases only daylight hours are included, and for practical reasons we define daylight for the model outputs as the time when the solar zenith angle is equal to or less than 89°. (The proper UNECE definition uses clear-sky global radiation exceeding 50 W m^{-2} to define daylight, whereas the EU AOT definitions use day hours from 08:00-20:00. Model outputs are also available using the EU definition, but not presented here).

The AOT40 levels reflect interest in long-term ozone exposure which is considered important for vegetation - critical levels of 3 000 ppb.h have been suggested for agricultural crops and natural vegetation, and 5 000 ppb.h for forests (Mills 2004).

1.3 Country Codes

Many tables and graphs in this report make use of codes to denote countries and regions in the EMEP area. Table 1.1 provides an overview of these codes and lists the countries and regions included in the present source-receptor calculations. Codes for natural sources (NAT) and boundary conditions (BIC) are discussed further in Chapter 6.

All Parties to the LRTAP Convention, except four, are included in the calculations. These are: Canada and United States of America, Monaco and Liechtenstein. The first two countries are not included because they lie outside the EMEP area domain. Monaco and Liechtenstein are not included because their emissions and geographical extents are below the accuracy of the source-receptor calculations.

Although Albania is not a Party to the LRTAP Convention, its situation in Europe and the extent of its estimated emissions justify a separate study of this country as emitter and receptor.

Malta is introduced as a receptor country. The estimated emissions from Malta are below the accuracy limits of the source-receptor calculations and do not justify a separate study of Malta as a emitter country.

1.4 Other Publications

This report is complemented with EMEP Status Report 4/2005 on Transboundary Particulate Matter in Europe and by country specific reports on the 2003 status of transboundary acidification, eutrophication, ground level ozone and PM.

In addition the following technical reports and a number of other reports and papers of relevance to transboundary air pollution and involving EMEP/MSC-W and CCC staff have become available in 2004/2005:

Peer-reviewed

J.E. Jonson, D. Simpson, H. Fagerli, and S. Solberg. Can we explain the trends in European ozone levels? *Atmos. Chem. and Physics*, 2005.

S. Tsyro. To what extend can aerosol water explain the discrepancy between model calculated and gravimetric PM_{10} and $PM_{2.5}$? *Atmos. Chem. Phys.* 5, 515-532, 2005 `http://www.copernicus.org/EGU/acp/acp/5/515/acp-5-515.htm`

W. Aas, J. E. Hanssen and J. Schaug. Field intercomparison of main components in air in EMEP. Submitted to *Water Air and Soil Pollution*

H. Fagerli and W. Aas. Modelled and observed trends of nitrogen in air and precipitation in Europe, 1980-2002. Submitted to *Water Air and Soil Pollution*

Code	Country/Region	Code	Country/Region
AL	Albania	HR	Croatia
AM	Armenia	HU	Hungary
AT	Austria	IE	Ireland
ATL	Remaining N.E. Atlantic	IS	Iceland
BA	Bosnia and Hercegovina	IT	Italy
BAS	Baltic Sea	KZ	Kazakhstan
BE	Belgium	LT	Lithuania
BG	Bulgaria	LU	Luxembourg
BIC	Boundary and Initial conditions	LV	Latvia
BLS	Black Sea	MD	Republic of Moldova
BY	Belarus	MED	Mediterranean Sea
CH	Switzerland	MK	The FYR of Macedonia
CS	Serbia and Montenegro	MT	Malta
CY	Cyprus	NAT	Natural+other emissions
CZ	Czech Republic	NL	Netherlands
DE	Germany	NO	Norway
DK	Denmark	NOS	North Sea
EE	Estonia	PL	Poland
EMC	EMEP Land Areas (all)	PT	Portugal
ES	Spain	REM	Remaining Land Areas
EU	European Community	RO	Romania
FI	Finland	RU	Russian Federation
FR	France	SE	Sweden
GB	United Kingdom	SI	Slovenia
GL	Greenland	SK	Slovakia
GE	Georgia	TR	Turkey
GR	Greece	UA	Ukraine

Table 1.1: Country/Region codes used in the source-receptor calculations

Russian Federation means the part of the Russian Federation inside the EMEP domain of calculations. The same applies to the Remaining N.E. Atlantic region and natural marine emission area. Remaining Land Areas refer to parts of North Africa and Asia within the model domain (REM=NOA+ASI). For North Africa this concerns parts of Morocco, Algeria, Tunisia, Libya and Egypt. With respect to Asia it includes Syria, Lebanon, Israel, parts of Uzbekistan, Turkmenistan, Iran, Iraq and Jordan. The European Union includes Austria, Belgium, Denmark, Finland, France, Germany, Greece, Ireland, Italy, Luxembourg, The Netherlands, Portugal, Spain, Sweden, United Kingdom, Cyprus, Czech Republic, Estonia, Hungary, Latvia, Lithuania, Malta, Poland, Slovakia and Slovenia.

T. Berg, W. Aas, J. Pacyna, H. T. Uggerud and M. Vadset. Atmospheric heavy metal concentrations at Norwegian background sites: Sources and temporal trends. Submitted to Symposium el lign til XIII International Conference on Heavy metals to the Environment

A. Lindskog, P.E. Karlsson, P. Grennfelt, S. Solberg, C. Forster. An exceptional ozone episode in northern Fenno-Scandinavia. 7th International Conference on Acid Deposition Acid Rain 2005. Submitted to *Water Air and Soil Pollution*

B. L. Skjelkvle, J. Stoddard, D. Jeffries, K. Trseth, T. Hgsen, J. Bowman, J. Mannio, D. Monteith, R. Mosello, M. Rogora, D. Rzychon, J. Vesely, J. Wieting, A. Wilander, A. Worsztynowicz. Regional scale evidence for improvements in surface water chemistry 1990-2001. In press, *Environmental Pollution*

S. Solberg, R. G. Derwent, Ø. Hov, J. Langner and A. Lindskog. European Abatement of Surface Ozone in a Global Perspective. *Ambio*, Vol. 34, No. 1, pp. 47-53, 2005

Other

L. Tarrasón, A. Benedictow, H. Fagerli, J.E. Jonson, H. Klein, M. van Loon, D. Simpson, S. Tsyro, V. Vestreng, C. Forster, A. Stohl, M. Amann and J. Cofala. EMEP Report 1/2004, Transboundary acidification, eutrophication and ground level ozone in Europe in 2003. Status Report 1/2005

V. Vestreng, K. Breivik, M. Adams, A. Wagner, J. Goodwin, O. Rozovskaya, and J. M Pacyna. Inventory review 2005. Emission data reported to LRTAP Convention and NEC Directive. Initial Review for HMs and POPs. EMEP/MSC-W Technical Report, 2005.

M. van Loon, L. Tarrasón, M. Posch. Modelling Base Cations in Europe EMEP/MSC-W Technical Report 2/2005

H. Klein, P. Wind, and M. van Loon. Transboundary Data by Main Pollutants (S,N,O3) and PM Country Reports. EMEP/MSC-W Note 1/2005

L. H. Slørdal, H. Laupsa, P. Wind and L. Tarrasonón. Local and regional description of Particulate Matter in the Oslo area. EMEP/MSC-W Technical Report 5/2004.

K. Tørseth (editor). Transboundary Particulate Matter in Europe: EMEP Status Report 4/2005

S. Solberg and A. Lindskog. The development of European surface ozone. Implications for a revised abatementpolicy A contribution from the EU research project NEPAP. EMEP/CCC-Report 1/2005

W. Aas. Workshop on the implementation of the EMEP monitoring strategy. EMEP/CCC-Report 2/2005

A.-G. Hjellbrekke. Data Report 2003 Acidifying and eutrophying compounds. EMEP/CCC-Report 3/2005

A.-G. Hjellbrekke and S. Solberg. Ozone measurements 2003. EMEP/CCC-Report 4/2005

J. Schaug. Measurements of particulate matter: Status report 2005. EMEP/CCC-Report 5/2005

W. Aas, A.-G. Hjellbrekke and J. Schaug. Data quality 2003, quality assurance, and field comparisons. EMEP/CCC-Report 6/2005

H. Uggerud and J. E. Skjelmoen. Analytical intercomparison of heavy metals in precipitation 2003 and 2004. EMEP/CCC-Report 7/2005

A.-G. Hjellbrekke, H. Uggerud, J E Hanssen and J Schaug. The twenty-second intercomparison of analytical methods within EMEP. EMEP/CCC-Report 8/2005

W. Aas and K. Breivik. Heavy metals and POP measurements, 2003. EMEP/CCC-Report 9/2005

S. Solberg, N. Schmidbauer and C. Dye. VOC measurements 2003. EMEP/CCC-Report 10/2005

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G. Mills. Mapping critical levels for vegetation. In UBA, editor, *UNECE Convention on Long-range Transboundary Air Pollution. Manual on Methodologies and Criteria for Mapping Critical Loads and Levels and Air Pollution Effects, Risks and Trends*. 2004. Constantly updated version available at `www.oekodata.com/icpmapping/`.

CHAPTER 2

Acidifying and eutrophying components, status in 2003

Hilde Fagerli

In this chapter we present model results for 2003 and compare them to measurements in the EMEP network. 2003 was a rather extreme year with respect to meteorological conditions, with a very warm and dry summer in large parts of central Europe. We discuss how the model results for 2003 for acidifying and eutrophying compounds differ from results for 2002, with special emphasis in the summer. Moreover, we show modelled depositions and use the depositions to different ecosystems to calculate exceedances of critical loads in Europe.

2.1 Comparison of observations and model results for 2003. Was 2003 a special year?

2003 has been characterized as a year with extreme weather conditions, due to the very warm and dry summer (Luterbacher et al. 2004). Both measurements and model calculations (Ordóñez et al. (2005), chapter 4) have demonstrated that this led to unusual high ozone concentrations. The effect on other atmospheric pollutants has not been subject to such detailed investigations. In this section we analyze the results for 2003 and discuss if the model is able to catch the pollution levels under such extreme conditions, focusing on the sulphur and nitrogen compounds.

2.1.1 Summary of model performance for 2003

In EMEP Status report I, part II (2003) we presented an extensive evaluation of the acidifying and eutrophying components for the years 1980, 1985, 1990 and 1995 to 2000. In EMEP Status report I, part III (2003) we showed a comparison of observations and modelled results for 2001. Last year we presented main results for 2002 with an updated EMEP Unified model, version 2.0. This version differed slightly from the 2003 version, as described in EMEP Status report 1/2004, however the main conclusions on the model performance remain the same. It has been shown that the EMEP model performance is rather homogeneous over the years, but depend on geographical coverage and quality of the measurement data.

The EMEP model has been validated for nitrogen compounds in Simpson et al. (2005) and for dry and wet deposition of sulphur, and wet depositions for nitrogen in Westling et al. (2005). This year we present results for 2003, calculated with the latest update of the EMEP Unified model, version 2.3. The emissions used for 2003 are documented in Vestreng et al. (2005). Ship emissions have been increased by 2.5 % per year, with 2000 as the reference year. Since last year, only changes with minor effect on the results for acidifying and eutrophying compounds have been introduced in the model. The previous evaluations of the model are still valid. Therefore, we refer to previous reports and papers for a detailed analysis of model performance.

Component	N_{stat}	Obs.	Mod.	Bias (%)	Corr.	Corr(d)
SO_2 (μg(S) m^{-3})	59	0.88	1.16	32	0.59	0.48
SO_4^{2-} (μg(S) m^{-3})	68	0.79	0.74	-5	0.71	0.62
NH_3+NH_4^+ (μg(N) m^{-3})	34	1.12	1.60	42	0.73	0.61
HNO_3 (μg(N) m^{-3})	14	0.13	0.11	-13	0.51	0.27
HNO_3+NO_3^- (μg(N) m^{-3})	35	0.48	0.59	22	0.88	0.71
NO_3^- (μg(N) m^{-3})	33	0.34	0.45	32	0.75	0.59
SO_4^{2-} wd (μg(S)m^{-2})	53	15274	15585	2	0.61	0.36
SO_4^{2-} cp (μg(S)l^{-1})	53	0.43	0.44	3	0.74	0.23
NH_4^+ wd (μg(N)m^{-2})	53	15315	11912	-22	0.73	0.35
NH_4^+ cp (μg(N)l^{-1})	53	0.42	0.34	-18	0.52	0.22
NO_3^- wd (μg(N)m^{-2})	53	12771	9139	-28	0.76	0.33
NO_3^- cp (μg(N)l^{-1})	53	0.36	0.26	-26	0.70	0.21
precip. (mm)	57	43468	46734	7	0.68	0.53

Table 2.1: Comparison of model results and observations for 2003. Annual averages over all EMEP sites with measurements more than 75 % of the days for air concentrations, and more than 25 % of the days for components in precipitation. N_{stat}= number of stations, wd=wet deposition, cp= concentration in precipitation, Corr.= spatial correlation coefficient, Corr(d)= temporal correlation between all daily observations and observations.

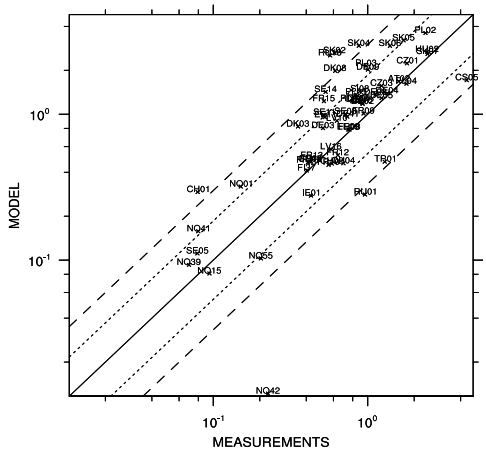
Component	N_{stat}	Obs.	Mod.	Bias (%)	Corr.	Corr(d)
SO_2 (μg(S) m^{-3})	68	0.85	1.06	24	0.56	0.46
SO_4^{2-} (μg(S) m^{-3})	70	0.74	0.67	-9	0.78	0.57
NH_3+NH_4^+ (μg(N) m^{-3})	42	1.20	1.63	35	0.72	0.58
HNO_3 (μg(N) m^{-3})	13	0.11	0.12	16	0.70	0.31
HNO_3+NO_3^- (μg(N) m^{-3})	41	0.48	0.59	23	0.90	0.68
NO_3^- (μg(N) m^{-3})	23	0.36	0.48	33	0.76	0.59
SO_4^{2-} wd (μg(S)m^{-2})	60	21935	20878	-4	0.65	0.32
SO_4^{2-} cp (μg(S)l^{-1})	60	0.50	0.44	-12	0.73	0.13
NH_4^+ wd (μg(N)m^{-2})	60	18735	15556	-16	0.74	0.39
NH_4^+ cp (μg(N)l^{-1})	60	0.39	0.31	-19	0.63	0.10
NO_3^- wd (μg(N)m^{-2})	60	16908	11468	-32	0.75	0.43
NO_3^- cp (μg(N)l^{-1})	60	0.36	0.24	-32	0.69	0.10
precip. (mm)	61	53215	58073	9	0.53	0.49

Table 2.2: Comparison of model results and observations for 2002. Annual averages over all EMEP sites with measurements more than 75 % of the days for air concentrations, and more than 25 % of the days for components in precipitation. N_{stat}= number of stations, wd=wet deposition, cp= concentration in precipitation, Corr.= spatial correlation coefficient, Corr(d)= temporal correlation between all daily observations and observations.

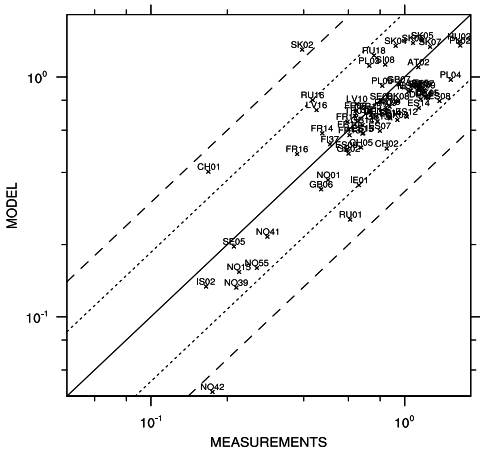
Sulphur dioxide in air

In Figure 2.1 we show scatter-plots for yearly averaged values for sulphur dioxide concentrations in air, 2003. The correlation coefficient between the yearly averaged observed and modelled data at EMEP stations is 0.59, which is relatively low compared to results for earlier years (see e.g. EMEP Status Report 1/2003, part II. Spatial correlation coefficients were normally between 0.7-0.8). Sulphur emissions have decreased substantially the last decades, especially in the high source areas. The emissions nowadays are more evenly distributed across Europe and concentration fields are more uniform. Thus, the model has to catch small differences in gradients to reproduce the spatial pattern. In addition, SO_2 is a primary pollutants, with large variation within small areas. This is rather difficult to reproduce in a regional model.

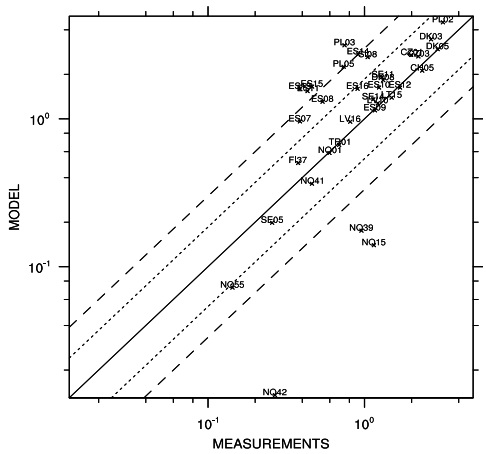
In general, the model overestimates the SO_2 concentrations, in average by 32 %. This is mainly caused by an overestimation in the winter time (and a somewhat smaller underestimation in summer), as can be seen from the time series in Figure 2.2. A possible explanation is that sesonal variation of the emissions, (based on data provided by the University of Stuttgart, IER, for 1990) have changed over time. Nowadays a larger part of emissions are released during the summer time, with the increasing use of air condition and more importantly the growth of telecommunications and computer hardware use.



(a) SO_2



(b) SO_4^{2-}



(c) NH_3+NH_4^+

Figure 2.1: Scatter-plots of modelled versus observed concentrations in air of sulphur dioxide, sulphate and sum of ammonia and ammonium (units: μg(S) $m^{-3}$and μg(N) m^{-3})

Sulphate in air

Scatter-plot for modelled versus measured sulphate concentrations in air for 2003 is presented in Figure 2.1. 97 % of the annual mean concentrations for the different sites are within a relative bias of 50% and approximately 93% within relative bias of 30%. The spatial correlation coefficient is high, 0.71. The daily correlations between model and measurements (not shown) are among the highest (r$\sim$0.6-0.8)for the acidifying and eutrophying compounds. This is partly due to sulphate being a secondary pollutant

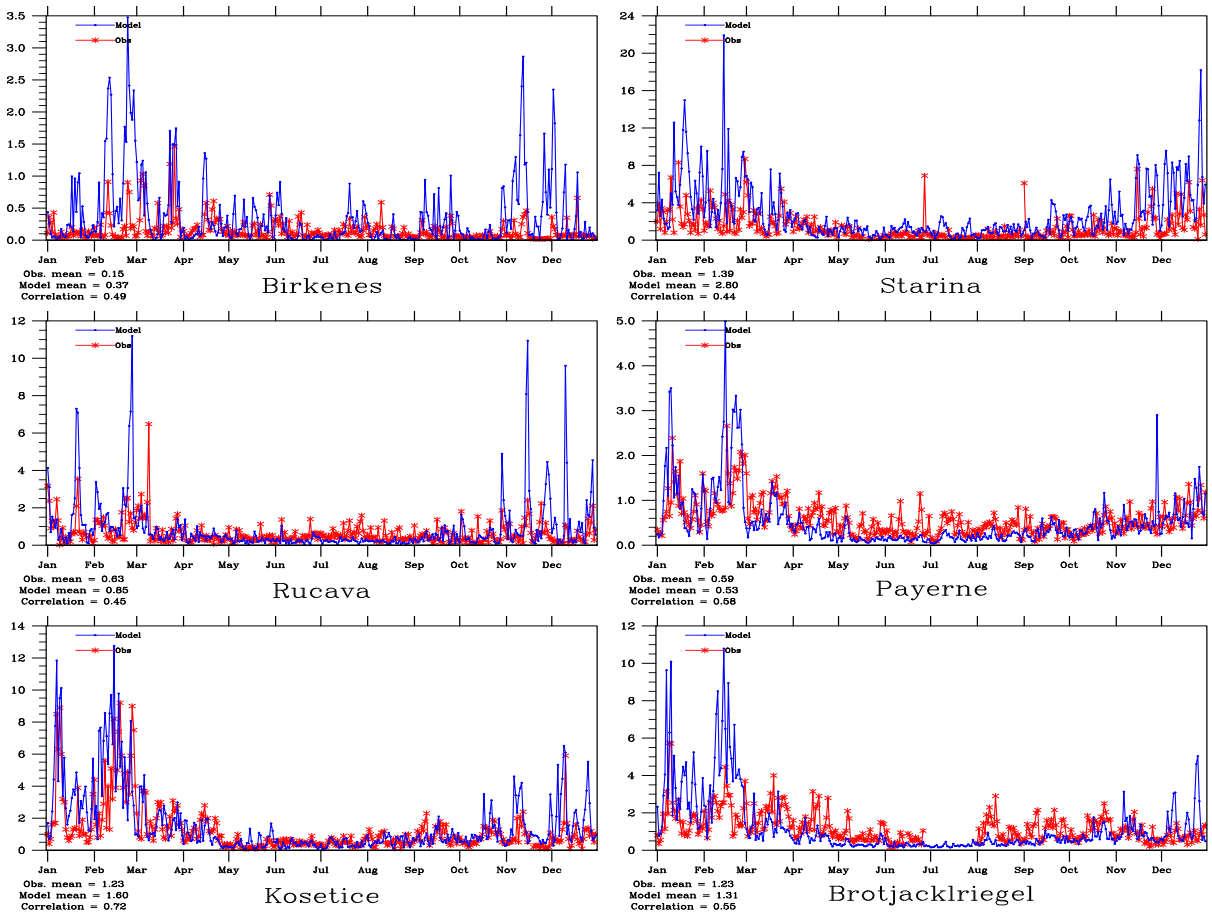


Figure 2.2: Daily time series of modelled versus observed SO_2 concentrations in air (units: μg(S) m^{-3}), 2003.

and thus less dependent on the ability of the model to simulate dispersion close to the sources. In addition the data quality of measurements of concentrations of SO_4^{2-} in air is among the best. Correlation coefficients and bias for sulphate in 2003 is similar to results for previous years. In Figure 2.3 we present daily time series for sites spanning the different concentration areas in Europe, from the high sulphate region in Eastern Europe to the low sulphate region around the Alps and in northern Europe. It is encouraging that the model capture both the absolute levels and the temporal variation at such different climatic and chemical conditions.

Nitrate and nitric acid in air

Measurements of airborne nitrate are expected to have a rather large uncertainty due to the very different physical characteristics of the compounds making up total nitrate. Whilst nitric acid is a spatially variable volatile gas with fast dry deposition, particulate nitrate dry deposits only slowly and hence concentrations are more determined by long

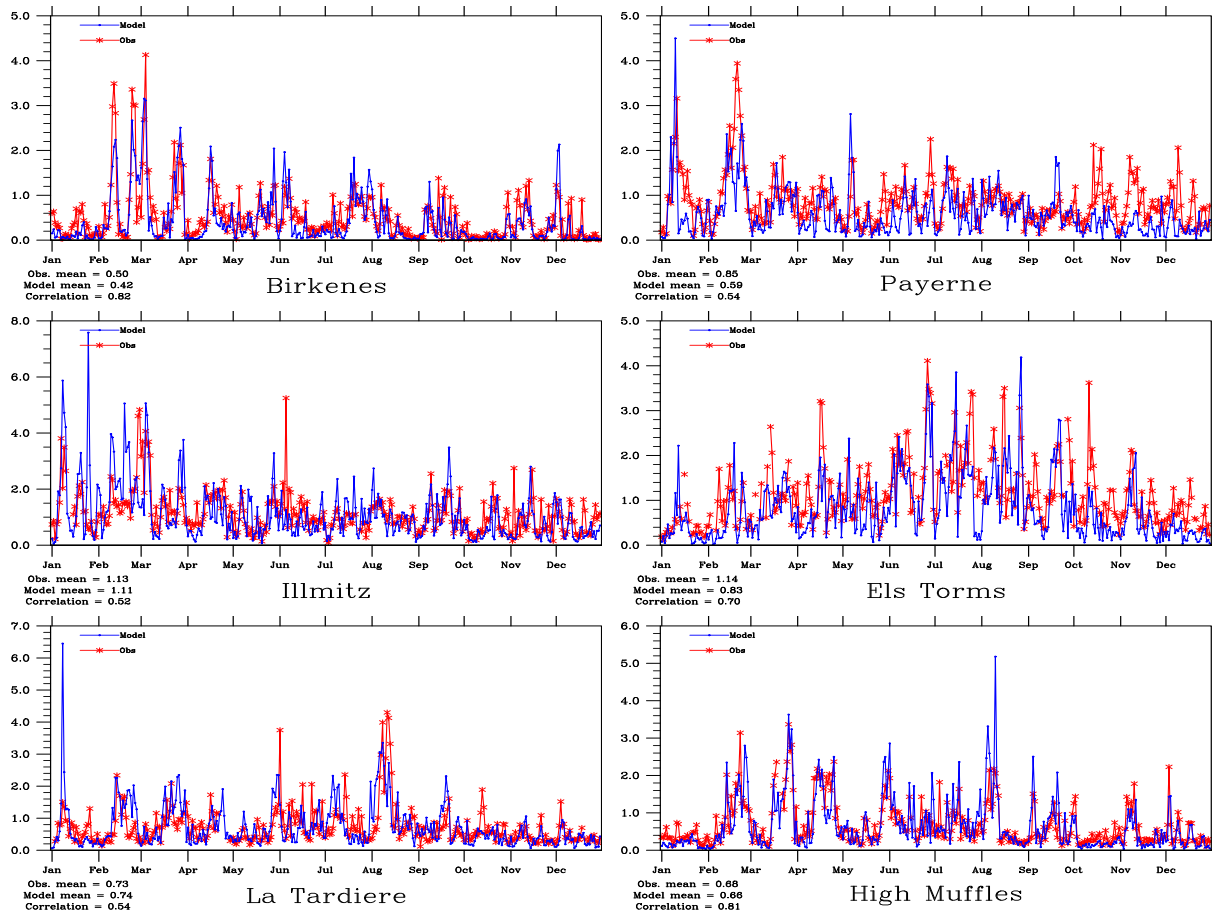


Figure 2.3: Daily time series of modelled versus observed SO_4^{2-} concentrations in air (units: μg(S) m^{-3}), 2003.

range transport.

In Figure 2.4 we show scatter-plots for total nitrate, particulate nitrate and nitric acid in air. Although there are some outlayers (LV10, LV16, SK02, PL03), the spatial correlation between model results and observations for total nitrate is as high as 0.88. The bias is 22%. The results for nitrate aerosol and nitric acid are somewhat worse, but this is expected as the monitoring data quality for these components are not as good as for total nitrate. The reason for this is that the individual concentrations of nitrate and nitric acid are biased when using the common filter-pack method (see EMEP Manual for sampling and chemical analysis, e.g www.emep.int). The sampling artifact is due to the volatile nature of ammonium nitrate, and separation of these gases and particles by a simple aerosol filter is unreliable. Better quality data can be obtained by using denuders, but this is a much more demanding method which few sites in the EMEP network are using. In Figure 2.5 we present daily time series for only some of the EMEP sites reporting total nitrate for 2003. In general, total nitrate is overestimated in the winter time. The reason for this is unclear, but it is possibly related to the

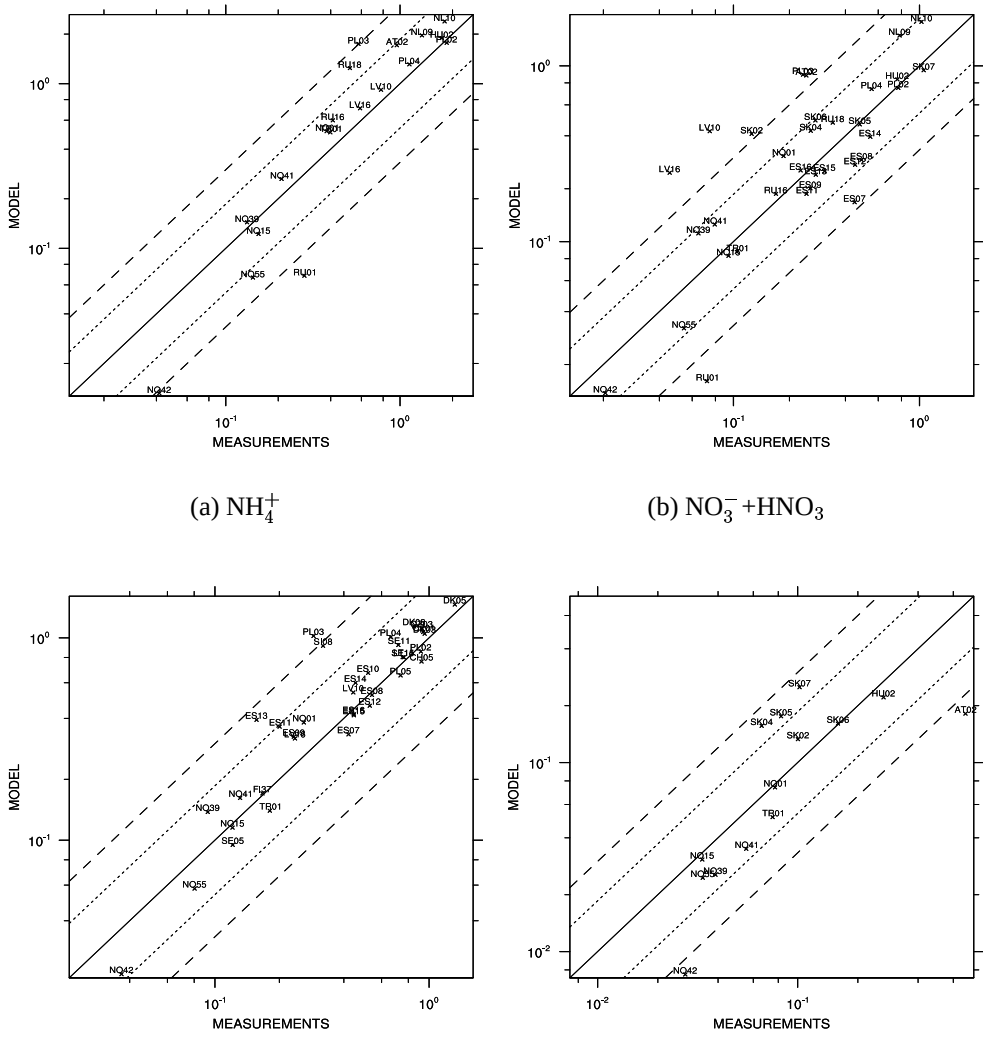
gas-aerosol partitioning. The replacement of the EMEP equilibrium chemistry with the EQSAM equilibrium module (results described in EMEP Status Report 1/2004) improved the agreement between the modelled and observed seasonal cycle, but there is still a systematic overestimation of nitrate in winter.

An increasing number of sites has reported measurements of nitric acid, and in Figure 2.6 we show comparison between model results and observations for some of these sites. Despite large uncertainties when using the filter pack method, the spatial correlation coefficient between model and measurements for these 14 sites is as high as 0.51, which is at the same level as for SO_2. There is no systematic over or underestimation. Temporal correlation coefficients range up to 0.57 (Birkenes), although most correlation coefficients are below 0.3. The low correlation between observations and model results is probably caused both by low quality of the measurements and deficiencies in the modelling of HNO_3. HNO_3 is among the most difficult nitrogen species to model as both production and loss reaction rates are uncertain (e.g. production via the reactions of NOx with OH and ozone and night time reaction of N_2O_5 on aerosols, loss via equlibrium reactions to ammonioum nitrate and very fast dry deposition). However, only Slovakia, Hungary, Norway, Austria and Turkey have reported nitric acid observations. More measurements, covering larger parts of Europe are needed in order to draw general conclusions.

Ammonia and ammonium aerosol in air

For 2003 as for the previous years, the rather limited number of sites (42 in 2003, 34 for more than 75 $\%$ of the days) that report measurements for NH_3+NH_4^+ (NH_x) are concentrated in the north with a few stations scattered around in the rest of Europe. The exception is Spain that reports measurements for 8 stations. In order to evaluate the model performance for NH_x properly, ammonia and ammonium should be studied separately. As for sum of nitrate, the individual concentrations of ammonia and ammonium are biased when using the common filter-pack method due to the volatile nature of ammonium nitrate. Further, very few measurements exist where the gaseous and particle phase are analyzed both separately and at the same time. Most sites that report ammonia are situated in Norway, and some of those are known to be strongly influence by local ammonia sources (W. Aas, CCC, pers. communication). Thus, we do no attempt to study model performance for ammonia as there is not enough unbiased data suitable for the analysis.

In Figure 2.4 scatter-plots for 2003 for model results versus observations for NH_x and aerosol ammonium are presented. The modelled yearly averages of the concentration of the sum of ammonia and ammonium in air are in good agreement with the monitoring data, whilst ammonium aerosols are somewhat overestimated (due to too high winter ammonium nitrate concentrations). In Figure 2.7 we present a selection of daily time series of ammonia plus ammonium for sites situated in different parts of Europe. In general, temporal correlation coefficients are high (0.5-0.7), although the model miss some of the high ammonia episodes in the spring (e.g. Campisabalos,



(a) NH_4^+ (b) NO_3^- +HNO_3

(c) NO_3^- (d) HNO_3

Figure 2.4: Scatter-plots of modelled versus observed concentrations in air of aerosol ammonium, sum of nitrate and nitric acid, aerosol nitrate and nitric acid (units: μg(N) m^{-3})

Osen) when agriculture activities peak. This may partly be explained by influence of local sources on the measurements.

Wet depositions

The ability of the model to predict concentrations in precipitations and wet depositions is limited by the accuracy of the precipitation fields used in the model. Scatter plots for modelled versus observed precipitation is shown in Figure 2.8. In average, the

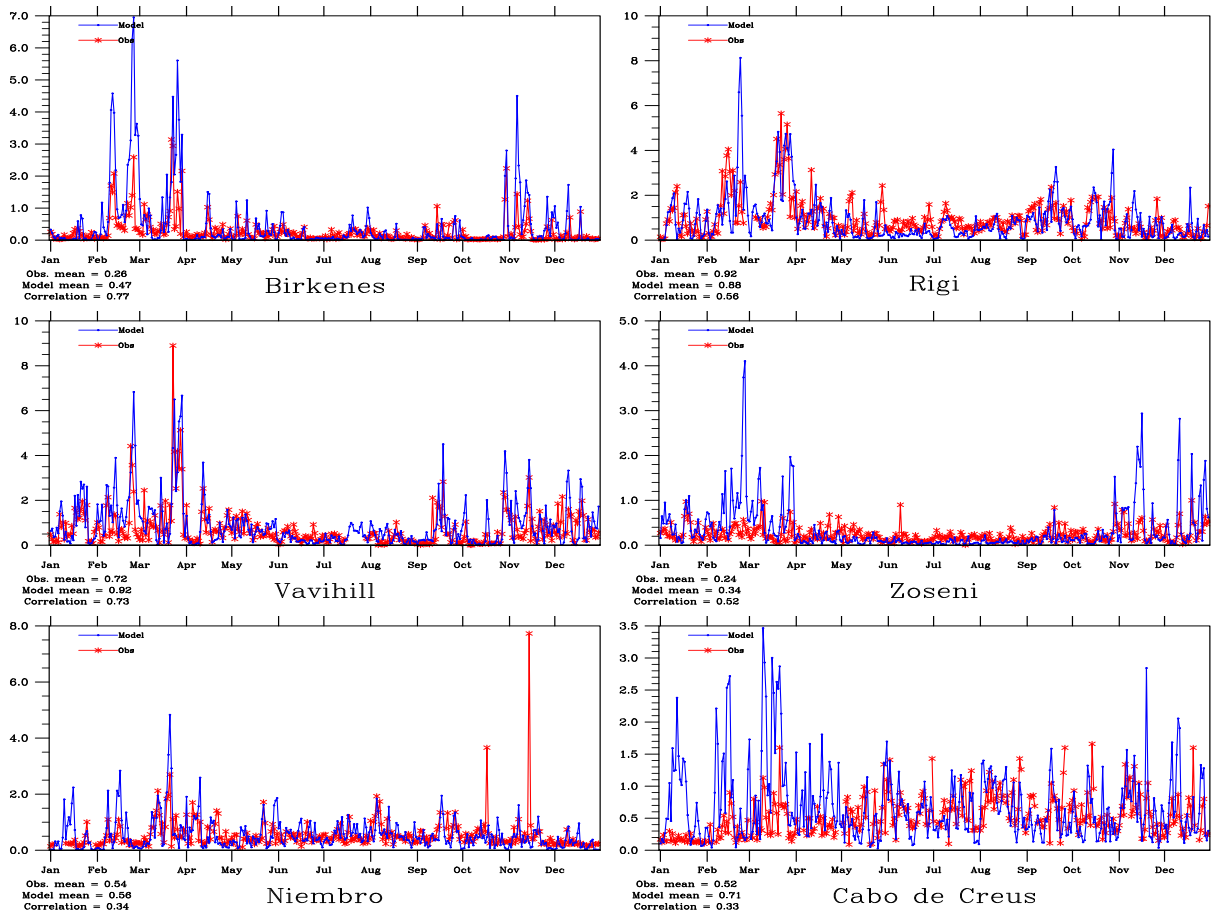


Figure 2.5: Daily time series of modelled versus observed total nitrate concentrations in air (units: μg(N) m^{-3}), 2003.

observed and modelled precipitation is very similar (bias=7 %). The spatial correlation is 0.68. In the same Figure, we show scatter-plots for concentrations in precipitation of sulphur, oxidized nitrogen and reduced nitrogen. The concentrations of nitrogen are in average somewhat underestimated (oxidized nitrogen -26 % and reduced nitrogen -18 %), whereas sulphur in precipitation is 3% overestimated. The spatial correlation coefficients range from 0.52 (reduced nitrogen) to 0.74 (sulphur).

In Westling et al. (2005) results of the EMEP model were compared with a completely independent data-set never before used in the evaluation or formulation of the model, that of the EU/ICP Forest (Level II) monitoring network. Modelled data from 1997 and 2000 were compared with observed deposition data from 160 ICP-Forest plots. In general, similarities between modelled and observed deposition in this study were reasonably good, despite the uncertainty in comparing measured plot data with modelled grid data. The EMEP model gave somewhat lower values for wet deposition in the whole deposition gradient compared to ICP, but differences in mean values were within 20% in 1997 and 30% in 2000. Modelled and observed concentrations of

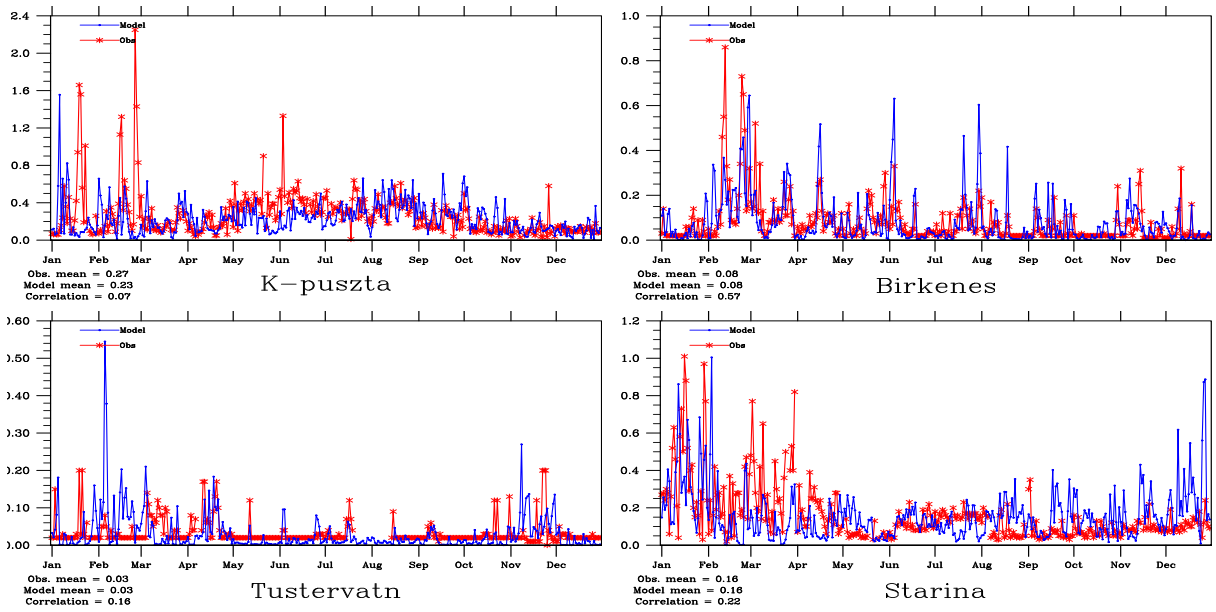


Figure 2.6: Daily time series of modelled versus observed nitric concentrations in air (units: μg(N) m^{-3}), 2003.

SO_4^{2-}, NO_3^- and NH_4^+ in precipitation were in average within 0-14% (underestimated), and the correlation between modelled and observed data rather high (correlation coefficient 0.7-0.9). Many of the sites showing large discrepancies between the EMEP model results and ICP data were found to have unusually high inter-annual variability in the precipitation amounts registered by ICP, suggesting that some differences may be due to sampling procedures or complex topographic effects. The overall conclusion was that the EMEP model performs rather well in reproducing patterns of S and N deposition to European forests.

The conclusions on model performance for nitrogen and sulphur in precipitation using the ICP data or the EMEP data are similar. However, the location of sites in the two networks are somewhat different. The ICP network is concentrated in central and north Europe, whilst the EMEP sites are more scattered across Europe. Further, the EMEP model has been shown to underestimate wet depositions in south Europe (EMEP Status Report I/2004). Thus, we cannot expect exactly the same results in the two studies.

It is, however, reassuring that the analysis of model performance using a completely independent data set gave similar conclusions as when using the EMEP data.

2.2 2003 versus 2002

As already mentioned, the summer of 2003 was extremely warm and dry in central and western Europe. Figure 2.9 show difference between mean surface (2 meters) temper-

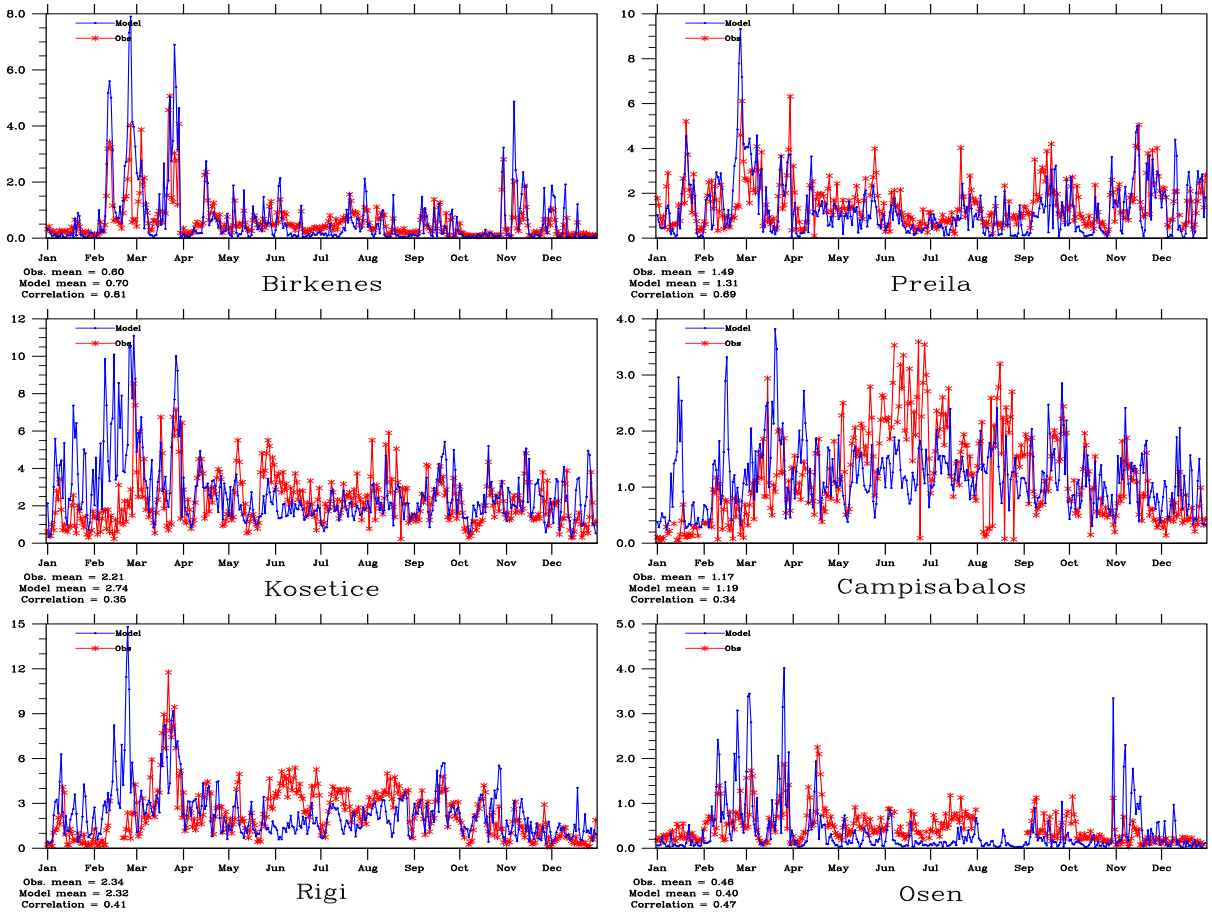
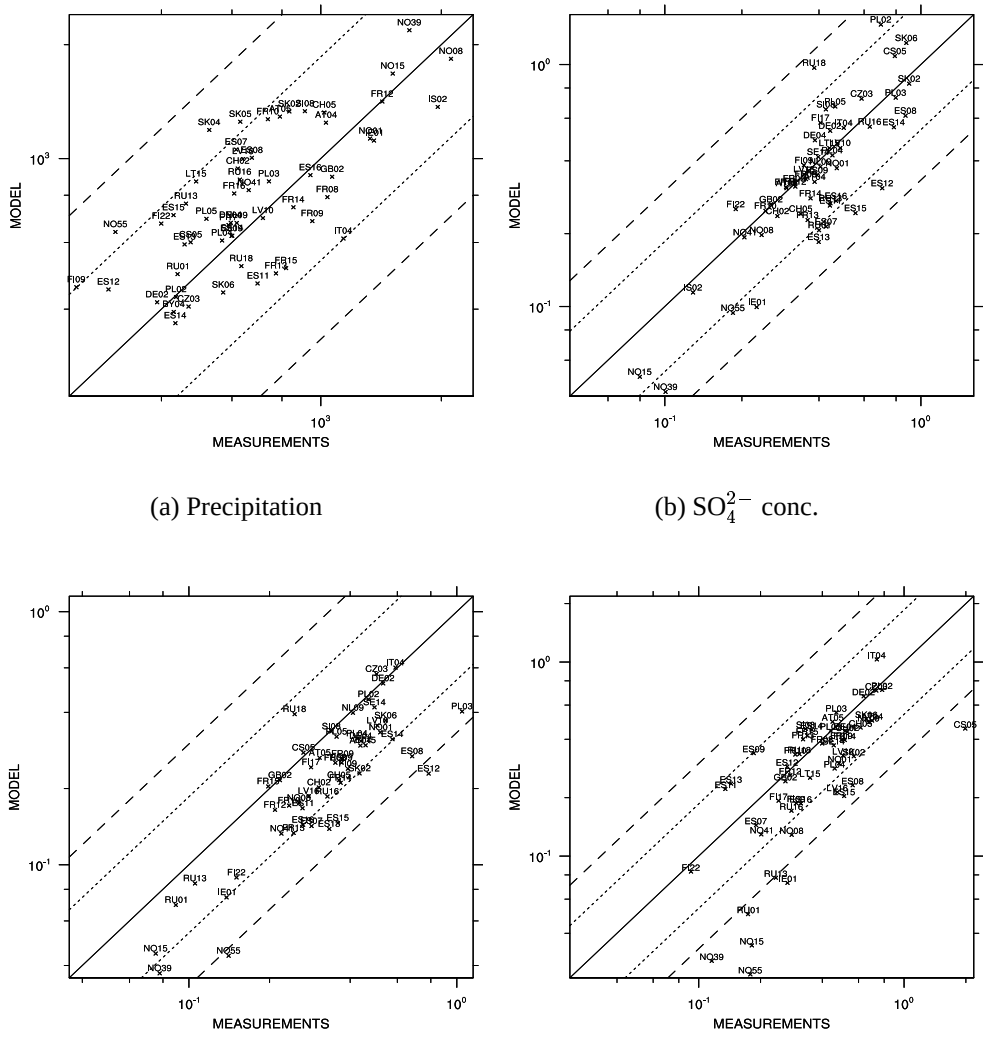


Figure 2.7: Daily time series of modelled versus observed NH_3+NH_4^+ concentrations in air (units: μg(N) m^{-3}), 2003.

ature in 2003 and 2002 for the whole year, Mars and August, respectively. France, Italy, Switzerland, Greece and countries along the eastern coast of the Adriatic sea had mean temperatures in August of 2-4 degree higher in 2003 than in 2002. Some regions (France, northern Spain, Switzerland and northern Italy) were more than 4 degrees warmer than in 2002. At the same time, the amount of precipitation was much lower in most of these regions, except parts of France. As a result, summer concentrations of the secondary inorganic particles (SIA) increased substantially (see Figure 2.10). For some regions, like for instance in Switzerland, summer concentrations of SIA were up to 60-80% higher than the previous year. As an example, we show in Figure 2.11 daily time series for 2002/2003 for two Swiss stations. Low cloud fractions lead to a lower amount of SO_2 to SO_4^{2-} oxidation and warm temperatures to a displacement in the equilibrium toward gaseous phase (at the cost of particulate ammonium and nitrate). However, for concentrations of particulates, it is the precipitation rate that has the largest influence. With practically no rain in the central/south of Europe in the end of July/beginning of August, the residence time of the aerosols in the atmosphere



(a) Precipitation

(b) SO_4^{2-} conc.

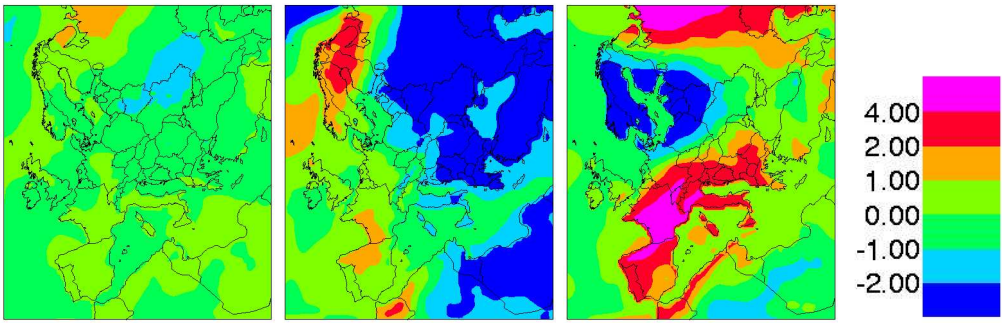
(c) NO_3^- conc.

(d) NH_4^+ conc.

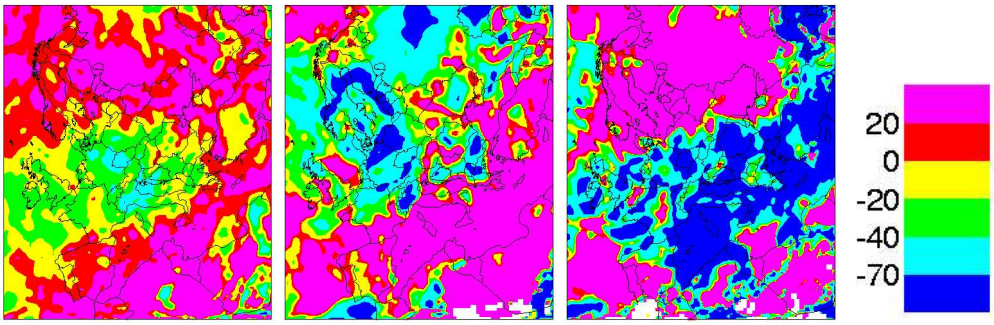
Figure 2.8: Scatter-plots of modelled versus observed results for concentrations of sulphate, oxidized nitrogen and reduced nitrogen in precipitation and accumulated precipitation (Units: mg(S)$l^{-1}$and mg(N)l^{-1})

increased dramatically. In the parts of France where the precipitation was equal to or lower than in 2002, concentrations of SIA decreased in 2003. Thus, the main reason for the high SIA concentrations in the summer 2003 was the low precipitation rate.

Concentrations of SIA vary around 20% (see chapter 5) from year to year due to inter annual variability in meteorological conditions. In 2003, concentrations of secondary inorganics were 10-40% higher than the previous year, but in southern parts of Europe up to 40-60% higher in August. However, the secondary inorganic con-

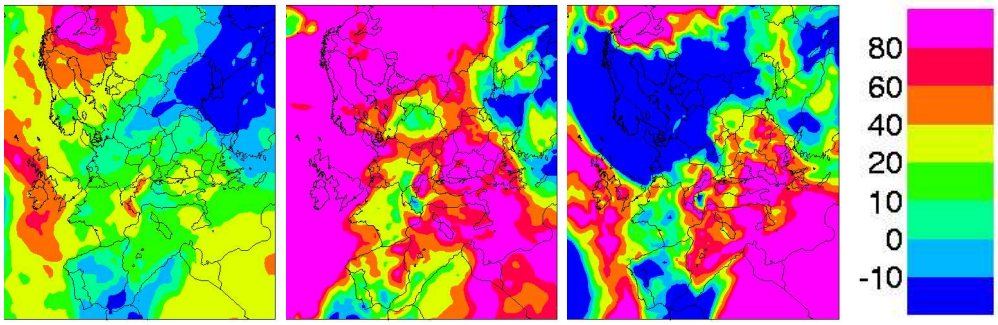


(a) Year (b) Mars (c) August



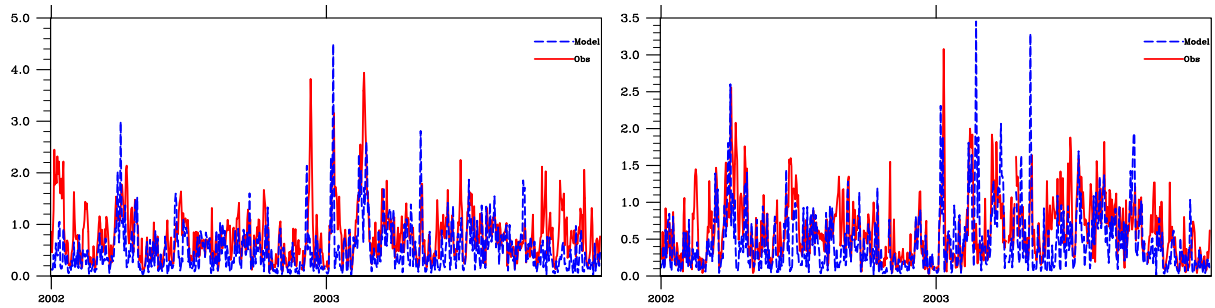
(e) Year (f) Mars (g) August

Figure 2.9: Difference in temperature (upper panel, units: Kelvin) and precipitation (lower panel, units: %) between 2003 and 2002 (relative to 2002).



(a) Year (b) Mars (c) August

Figure 2.10: Difference in concentrations of SIA (=sulphate+ammonium+nitrate) (units: %) between 2003 and 2002.



(a) Payerne (b) Rigi

Figure 2.11: Daily time series of sulphate concentrations in air (units: μg(S) m^{-3}) for 2002-2003 at two Swiss stations.

centrations tend to be highest in the winter season (see chapter 3), thus an increase in summer concentration does not have a large impact on the yearly average. Further, in February and Mars 2003, large parts of central and north Europe received very little precipitation. As a consequence the concentrations of SIA were much higher in this season than the years before.

In Figure 2.12 we present time series for SO_2 and sulphate averaged over the three summer months (June, July, August) from 1997 to 2003 for stations situation in central-south Europe having continuous measurements from 1997 to 2002 (Czech republic, Slovakia, Slovenia, Austria, France, southern Germany, Hungary). From these plots it is clear that in average 2003 concentrations of SO_2 and SO_4^{2-} was high, but not extreme with respect to the other years. It should be noted, however, that emissions have decreased during this period and the effect from emissions are not separated out in these plots.

In Figure 2.13 we present trends (1995-2003) in sulphate (monthly concentrations) averaged over stations in different parts of Europe. For central-west Europe (including 3 Swiss stations, which are the only EMEP stations that has reported measurement for the whole period) the sulphate concentrations in the summer 2003 was higher than what has been observed since 1996, but in other parts of Europe the 2003 summer is not an outlayer. In contrast, the winter (February, Mars) concentrations is among the highest that has been observed since emissions were on a much higher level. Very few sites in south Europe report measurements of ammonium plus ammonia or total nitrate in air, thus the same analysis is difficult to perform for these species. Model calculations indicates, however, that in the same way as for sulphate, the summer concentrations were high, but not higher than within the inter annual variability that has been observed before. Summer concentrations of nitrate and ammonium was 20-30% higher in 2003 than in 2002 in southern Europe.

Inter annual meteorological variability is large and becoming larger (see chapter 5). We conclude that although concentrations of the inorganic compounds were somewhat

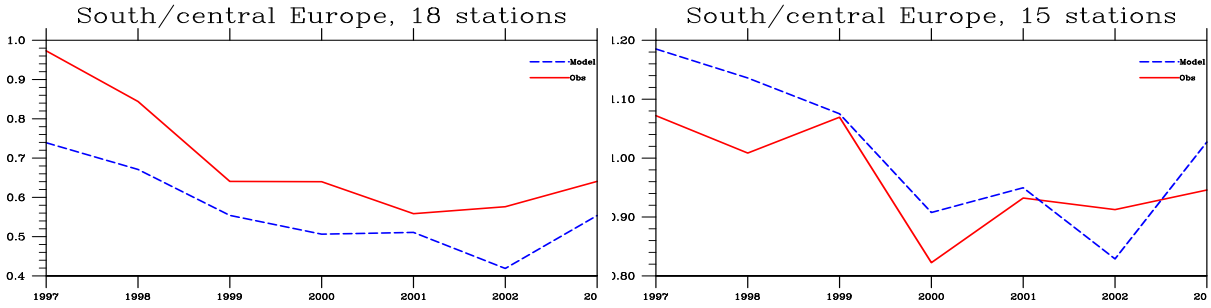


Figure 2.12: Monthly timeseries averaged over sites in areas that experienced extreme temperatures in the summer 2003. SO_2 in air (left) (units: μg(S) m^{-3}) and sulphate in air (units: μg(S) m^{-3}) (right).

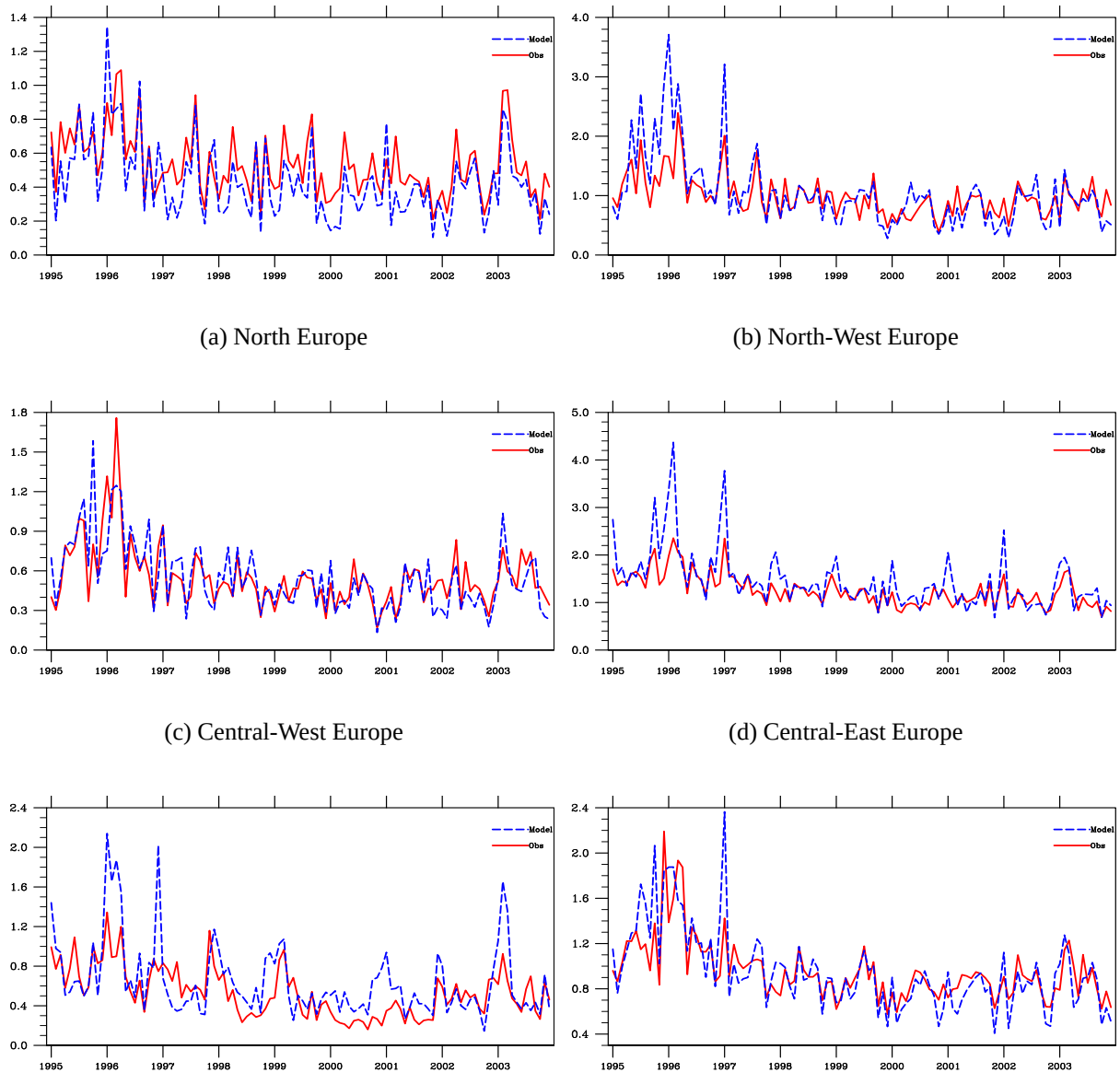
high in the summer 2003, they are not extreme with respect to what has been observed the latest years. Going back to 1996 and earlier, the emissions were much higher, and the effect of meteorological variability smaller than the effect of emissions - thus in general higher concentrations were observed.

2.2.1 Can the model tackle extreme conditions?

In order to investigate the model performance of the EMEP Unified model under extreme weather conditions like the ones in 2003, we have compared the model performance at the stations in central Europe for the summer 2003 to summer 2002. In Table 2.3 we show model and measured averages, bias and spatial correlation for the collection of sites that reported measurements for both years. For all components in air, concentrations in the summer 2003 are higher than in 2002 both in the model and in the measurements. Further, wet depositions of sulphur and nitrogen are significantly lower in 2003 than in 2002. The spatial correlation between observation and model for the wet depositions are high (0.7-0.8) and approximately at the same size in both years. For oxidized nitrogen in precipitation the correlation is much lower in 2003, but this is mainly due to one outlyer, the Italian station Montelibretti. For this stations, the model underestimates the precipitation largely in 2003 and as a consequence also the wet depositions, although most severely for nitrate.

For air concentrations, the results are more mixed. SO_2 is less underestimated in the summer of 2003, but the spatial correlation decrease from 0.59 to 0.23. The Slovakian stations are significantly more overestimated in summer 2003 whilst some French stations (FR10, FR16) are more underestimated. The reason is unclear as both areas experienced approximately the same meteorological conditions. The method used at the French stations has a very high detection limit, and thus less suitable with the low concentrations nowadays. At the Slovakian sites there are no appearent problems with the measurement data.

For nitrate, more than half of the stations are Spanish. In the summer of 2003, the



(a) North Europe

(b) North-West Europe

(c) Central-West Europe

(d) Central-East Europe

(e) East Europe

(f) South Europe

Figure 2.13: Monthly time series for sulphate in air, 1995-2003 (units: μg(S) m^{-3}). Comparison of model results and observations in different regions in Europe. Only sites with continious measurements are included.

		2003				2002			
Component	Stations	Obs.	Mod.	Bias	Corr.	Obs.	Mod.	Bias	Corr
SO_2 (μg(S) m^{-3})	24	0.64	0.52	-18	0.23	0.60	0.42	-30	0.59
SO_4^{2-} (μg(S) m^{-3})	30	1.11	1.07	-2	0.72	1.05	0.95	-9	0.87
NH_3+NH_4^+ (μg(N) m^{-3})	15	1.48	1.85	25	0.32	1.21	1.71	41	0.24
HNO_3+NO_3^- (μg(N) m^{-3})	15	0.55	0.44	-19	0.12	0.40	0.42	4	0.01
SO_4^{2-} wdep (μg(S)m^{-2})	32	2970	2110	-28	0.63	4824	2887	-40	0.57
NH_4^+ wdep (μg(N)m^{-2})	32	3263	2113	-35	0.79	3846	2839	-26	0.76
NO_3^- wdep (μg(N)m^{-2})	32	2441	1369	-43	0.39	2808	1731	-38	0.76
precipitation (mm)	32	4511	5022	11	0.84	8014	8594	7	0.83

Table 2.3: Results for summer 2003 and 2002

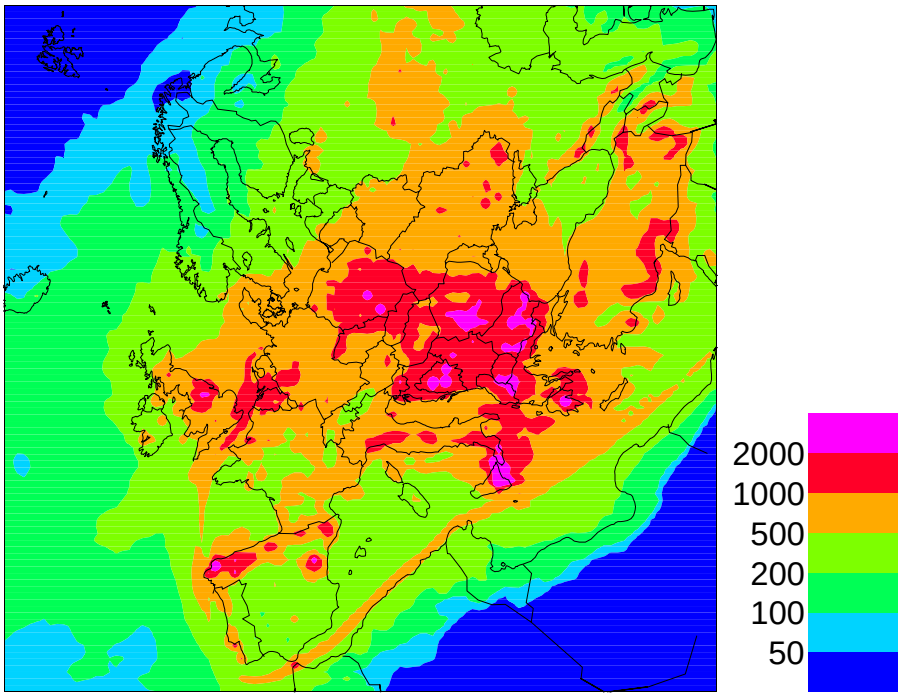
modelled absolute levels of these stations are in better agreement with the observations (less overestimated than in 2002), whereas the Czech and a German station are underestimated with approximately the same amount as in 2002. In general, the correlation between observed and measured total nitrate is low in the summer in the region we study here, possibly due to measurement artefacts using the filterpack method. It is the same stations that measure total nitrate and the sum of ammonia and ammonium, and the same considerations applies to the results for reduced nitrogen in air. The precipitation is somewhat more overestimated at the Spanish sites in 2003, which explains the relatively lower air concentrations.

Because there are a limited number of sites that measure total nitrate and ammonia+ammonium, and most of them concentrated in one area (Spain), it is difficult to draw general conclusions. It seems however, that the model performance is strongly connected to the quality of the precipitation fields. For sulphate, measurement stations from a wider region is available, and model performance (here quantified by bias and spatial correlation) for both years are rather similar.

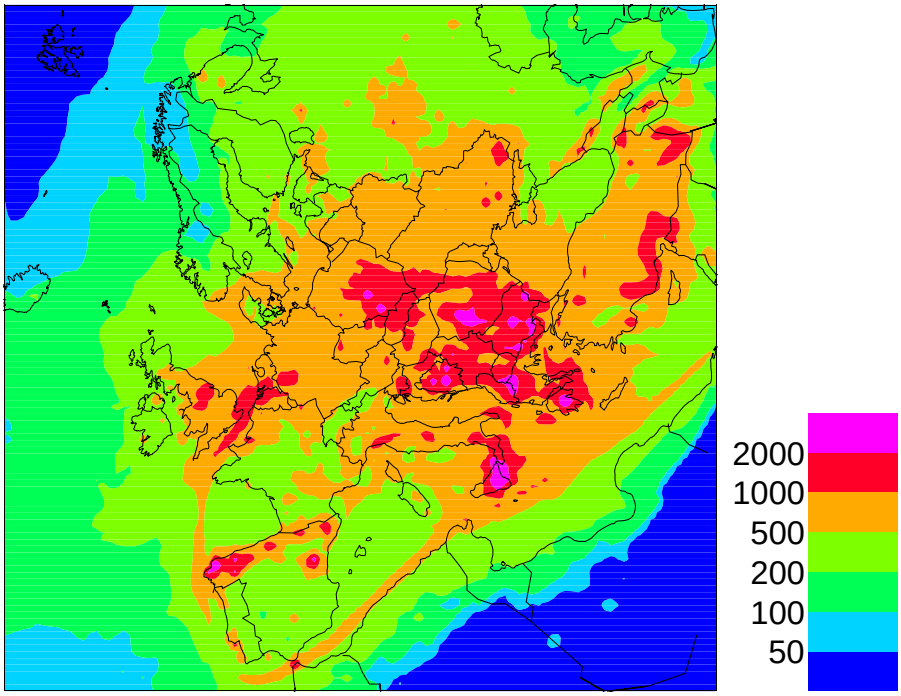
We conclude that the model performance for the sulphur and nitrogen compounds are at the same level in the extreme summer of 2003 as in 2002.

2.3 Deposition of sulphur and nitrogen in 2003 and exceedances of critical loads

In Figure 2.14, 2.15 and 2.16 we present maps of depositions for 2002 and 2003 for oxidized sulphur, oxidized nitrogen and reduced nitrogen. The maps presented here for 2002 differ from the ones presented last year in two ways; 1) a slightly different model version was used last year and 2) the gridding of the emissions have been slightly revised. However, these changes do not affect the model results significantly. To secure consistency with the 2003 calculation, we present here results calculated with the same model version (rv 2.3) and the same method for gridding of the emissions. In general, sulphur depositions are somewhat lower in eastern Europe in 2003 than in 2002 due to lower emissions in this region. For NO_x, emissions in 2003 are almost at the same

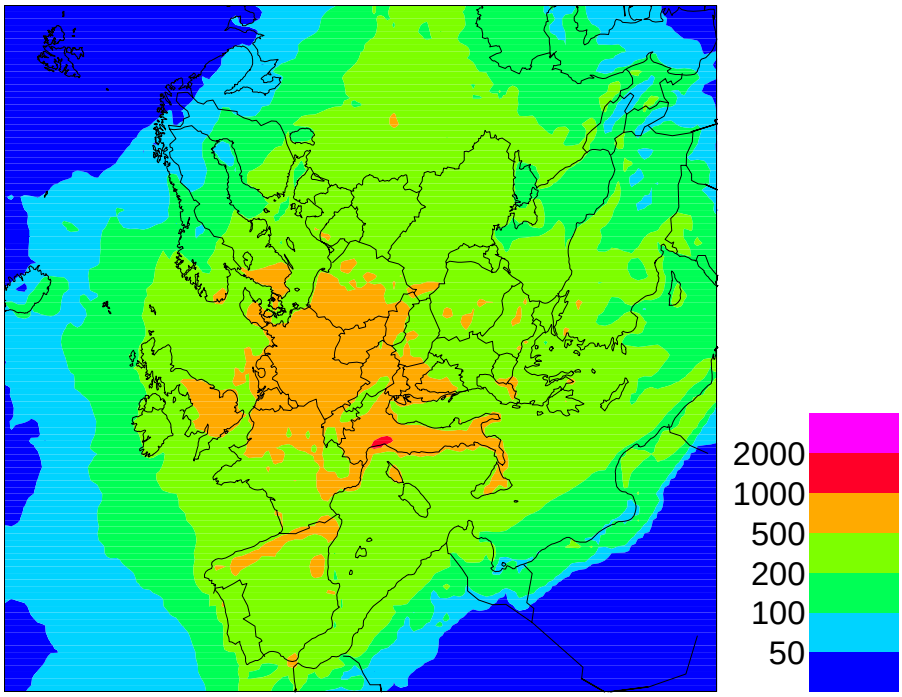


(a) 2002

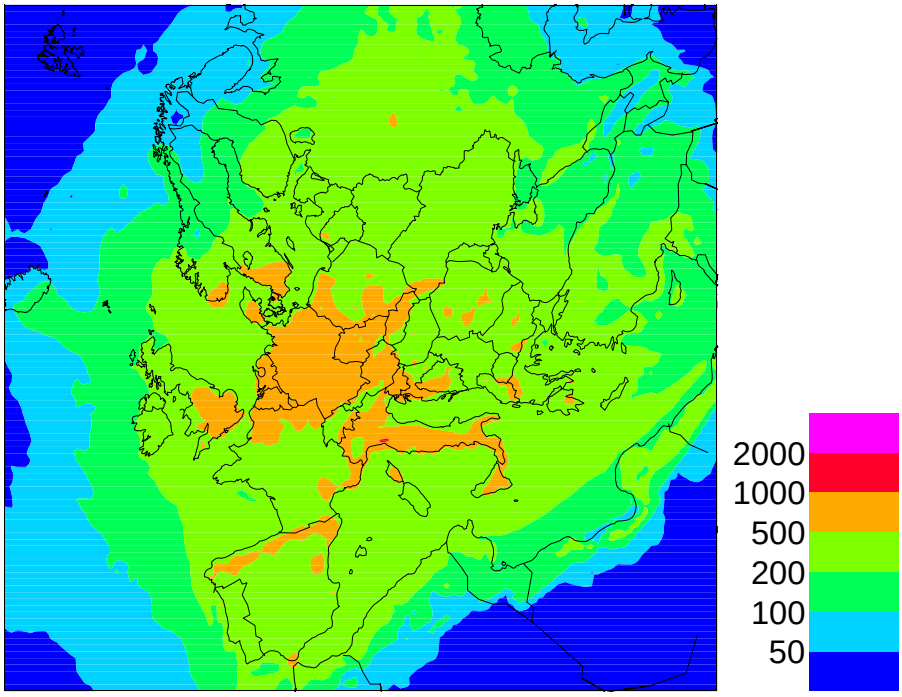


(b) 2003

Figure 2.14: Modelled deposition of sulphur (units: mg(S)m^{-2}) for 2002 and 2003.

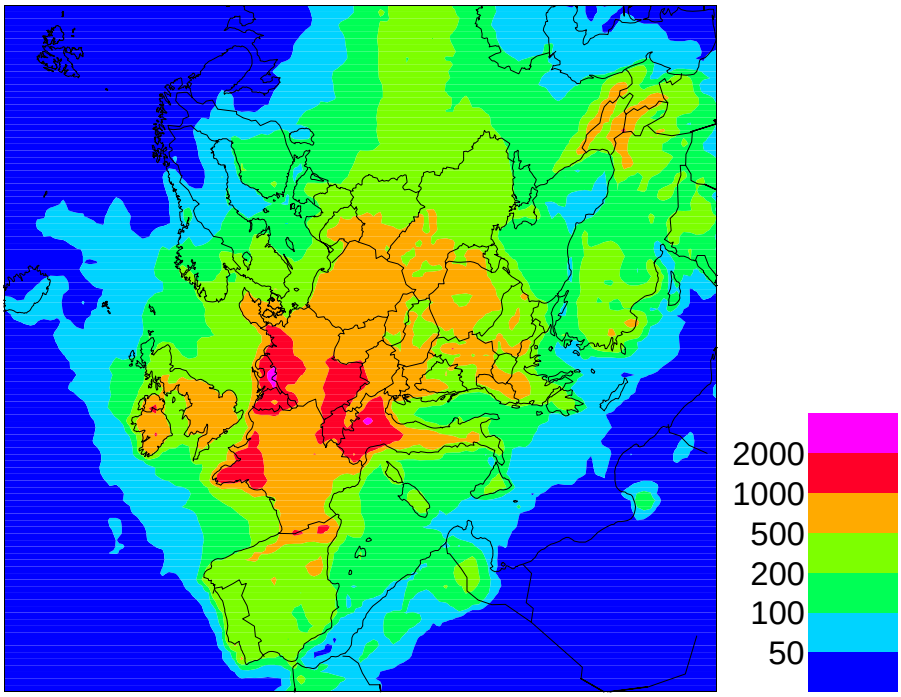


(a) 2002

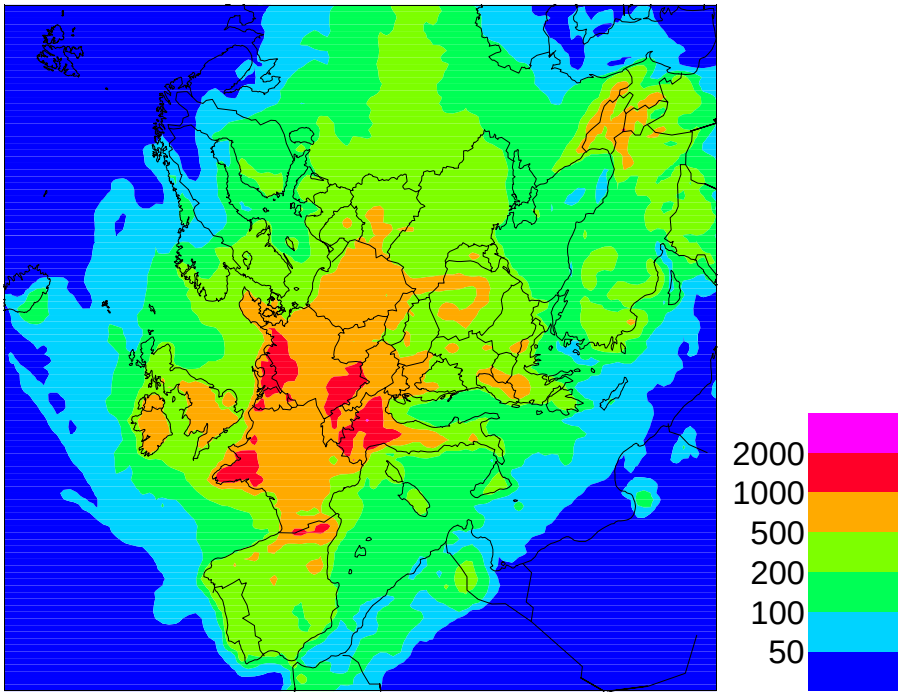


(b) 2003

Figure 2.15: Modelled deposition of oxidized nitrogen (units: mg(N)m^{-2}) for 2002 and 2003.



(a) 2002



(b) 2003

Figure 2.16: Modelled deposition of reduced nitrogen (units: mg(N)m^{-2}) for 2002 and 2003.

level as in 2002, and there are no major changes in the deposition pattern. Poland and France receive somewhat less deposition of oxidized nitrogen due to reduction of emissions, but otherwise depositions of oxidized nitrogen are very similar in the two years. Lower reduced nitrogen depositions are calculated in Germany, Netherlands, Switzerland and Italy. All of them, except Switzerland, have reported lower ammonia emissions in 2003 than in 2002.

These deposition fields have been used to calculate the exceedances of critical loads in Europe.

Two different critical loads has been used: the critical load of nutrient nitrogen, the maximum acceptable deposition of nitrogen not causing ethrophication of ecosystems, and critical loads of acidity, the maximum deposition of sulphur and nitrogen not causing detrimental leaching of acidity. The calculated exceedances of the critical loads are the so-called average accumulated exceedances, described e.g in Posch et al. (2001). Critical load data is documented in Posch et al. (2005).

The modelled ecosystem specific deposition data are used to calculate exceedances of critical loads for acidification and nutrient nitrogen. The dry deposition scheme used in the model distinguishes between a number of land use categories. Dry deposition will thus differ for different land use types. This has implications for the exceedances of critical loads. Dry deposition is added to the wet deposition (unaffected by land use) to get the total deposition.

The modelled ecosystem specific deposition data are used to calculate exceedances of critical loads for acidification and nutrient nitrogen. Three ecosystem classes have been identified as most significant for the calculation of exceedance to critical loads. These are:

- Forest ecosystems
- Semi-natural vegetation
- Surface waters

For forest ecosystems we use the average of depositions to coniferous and deciduous forest. Forest waters depositions are approximated by the grid average dry depositions. In all cases, we use the grid average for the wet deposition.

Forests receive more deposition than most other land use types. Thus depositions to forests per unit-area tend to be higher than levels calculated for the grid average. Therefore, exceedances of critical loads calculated using ecosystem specific depositions instead of the grid averaged depositions gives a higher estimate of exceedances to critical loads, as was shown in EMEP status report 1/2004. This has to be kept in mind when comparing maps of exceedances of critical loads with maps presented for previous (to 2004) years in earlier reports.

In Figure 2.3 we present maps of exceedances to critical loads for 2002 and 2003. Both nutrient and acidity critical loads are less exceeded in 2003 than in 2002. For exceedances of acidity critical loads, the most striking differences are in Poland were

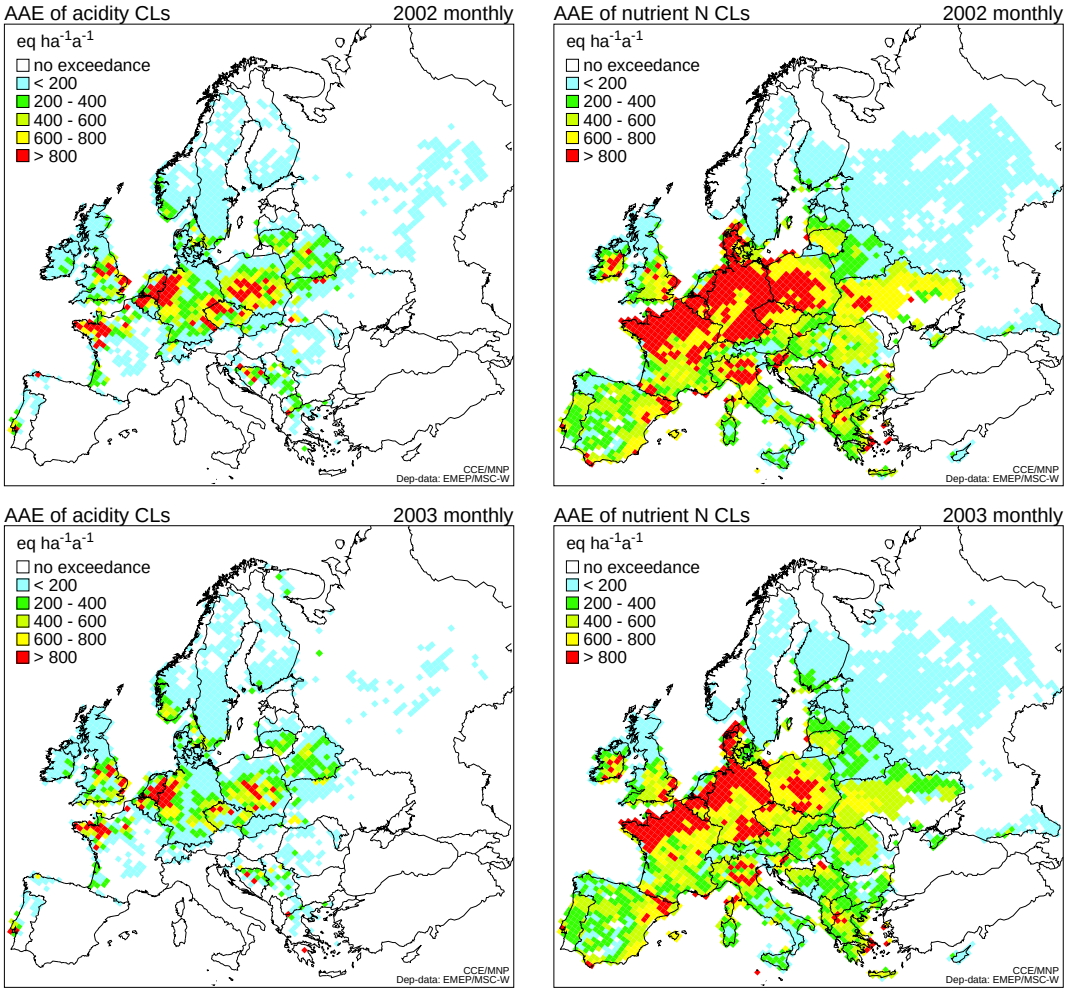


Figure 2.17: Calculated exceedances of critical loads in 2003 and 2002 (eq/ha/yr). Left column: average accumulated exceedance of acidity, right column: average accumulated exceedance of nutrient. Source: CCE

the exceedance in 2003 is much lower than in 2002. Small improvements are also made in Switzerland, southern Norway and Belarus. However, the largest changes are seen for nutrient exceedances. Especially in Germany, France, Poland, southern England and northern Italy the situation has improved. The emissions for these countries were similar in the two years, but due to the dry conditions in central Europe in 2003, less nitrogen was wet deposited and more pollution were transported out of this region (and into e.g Norway, Sweden and Finland). Consequently, the critical loads were less exceeded. In the nordic countries, the opposite situation occured - this region received more deposition in 2003 compared to 2002 and the critical loads were more exceeded.

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CHAPTER 3

Particulate Matter concentrations, status in 2003

Svetlana Tsyro

In this chapter, we study the effect of meteorological variability on the European Particulate Matter concentrations based on the analyses of the 2003 air pollution situation. The year 2003 was characterised by an exceptionally hot and arid summer in western, central and southern Europe. In particular, it was very little precipitation which affects most the PM concentrations in those areas. The main differences between aerosol concentration fields in 2003 and 2002 are discussed in terms of changes in emissions and in meteorological conditions. Finally, we compare model PM results with observations at the EMEP monitoring network, with particular focus on evaluating the model performance in the year 2003 versus 2001 and 2002.

3.1 Model results for PM

3.1.1 Concentration fields in 2003

Calculations of particulate matter concentrations in 2003 presented in this chapter have been performed with the EMEP Unified model (UNI-ACID model version). The recent model development with respect to the calculation of aerosol processes is described in Tsyro (2005a).

In these model calculations we have used 2003 meteorology calculated with the PARLAM_PS weather prediction model and emissions of SO_2, NO_x, NH_3, $PM_{2.5}$ and PM_{10}in 2003 (Vestreng, 2005). Grid segregation and sector allocation of the national total emissions has been done as described in Tarrasón et al. (2004). The chemical speciation of primary PM emissions is based on the IIASA preliminary inventory of submicron carbonaceous particle emissions (Kupiainen and Klimont (2004)). According to these estimates, the emissions of primary $PM_{2.5}$ have been divided into organic

carbon (OC), elemental carbon (EC) and inorganic components, assumed to be mineral dust. Primary coarse PM has been divided into elemental carbon and mineral dust only, using a very preliminary emission estimate of coarse EC (Klimont, personal communications). Emissions of primary anthropogenic coarse OC are not available at present. Natural aerosol emissions included in the model are sea salt spray and dust emissions by wind erosion. The calculation scheme for sea salt aerosol production has been improved since last year. A newly developed parameterisation for wind-blown mineral dust emissions from deserts and agricultural fields have been implemented for testing in the Unified model. Model details on the modelling of natural particle sources can be found in Tsyro (2005a). At present, the model calculates 7 individual aerosol components: SO_4^{2-}, NO_3^-, NH_4^+, OC, EC, mineral dust and sea salt. Also, particle water content is calculated for temperature 20 $^\circ$C and relative humidity 50%, which are equilibrating conditions required for gravimetric PM measurements (Tsyro (2005b)).

Annual mean concentrations of PM_{10} and $PM_{2.5}$ calculated with the EMEP Unified model for the conditions of 2003 are presented in Figure 3.1. The calculated PM_{10} and $PM_{2.5}$ concentrations include primary PM_{10}, secondary inorganic aerosols (SIA), namely SO_4^{2-}, NO_3^- and NH_4^+, sea salt, wind-blown mineral dust and particle water. Compared with calculations performed in the previous years, accounting for the contribution of natural sources of mineral dust to PM has increased $PM_{2.5}$ and PM_{10} concentrations. The influence of natural dust is especially pronounced in Mediterranean countries and the southern parts of East Europe and Russia. Accounting for particle water further increases the concentrations of $PM_{2.5}$ and PM_{10} by about 10-25% on average.

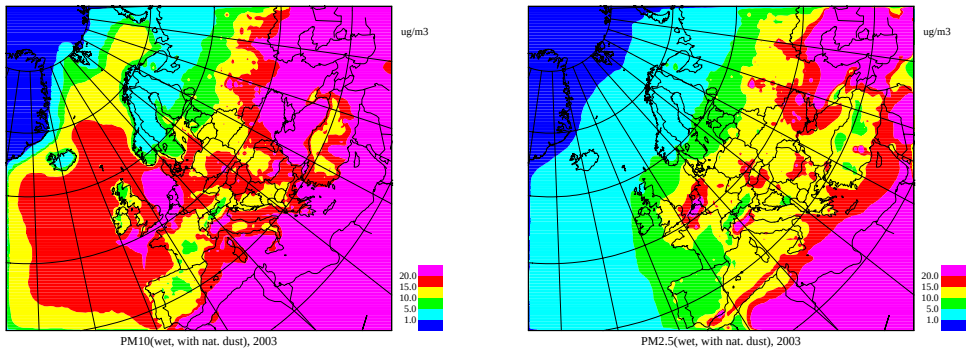

Figure 3.1: Model calculated PM_{10} and $PM_{2.5}$ in 2003 (particle water and wind-blow dust included). Units: μg/m^3.

Figure 3.2 shows the annual mean concentrations of primary anthropogenic $PM_{2.5}$ and coarse PM, secondary inorganic aerosols (SIA) and natural PM_{10} in 2003. According to our calculations, SIA is the largest component in $PM_{2.5}$ and PM_{10} concentrations in most EMEP area. Primary anthropogenic PM becomes more important in big cities (e.g. Paris, Moscow, Istanbul) and industrial regions with large sources of primary PM emission (e.g. north of Kola Peninsula, eastern Ukraine, the Ural area) (Figure 3.3,

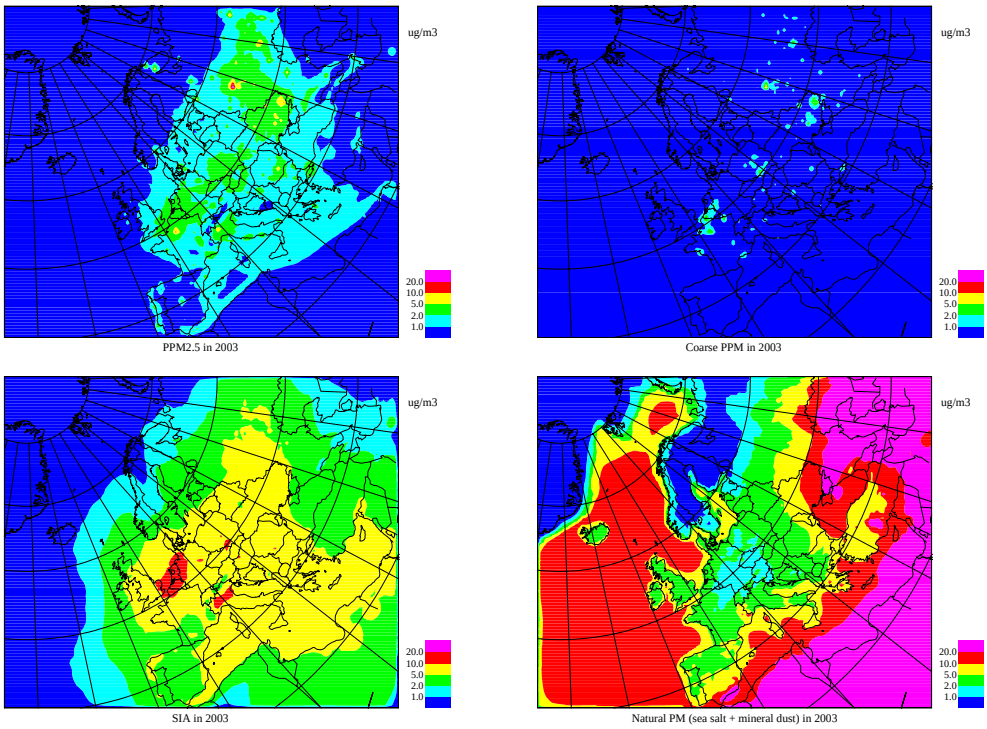

Figure 3.2: Model calculated concentrations of primary $PM_{2.5}$ (upper left), primary coarse PM (upper right), SIA (lower left) and natural particles (sea salt and wind-blown mineral dust) (lower right) in 2003. Units: $\mu g/m^3$.

left map).

Initial model calculations show that the concentrations of aerosols from natural sources, which are sea salt spray and soil wind erosion in the model, are 0.5-5 $\mu g/m^3$ over most Europe (not shown here; see Tsyro (2005a)). They reach 10-20 $\mu g/m^3$ in the south and south-east of EMEP domain due to the effect of wind-blown dust from the deserts (Sahara, Kara Kum, Kyzyl Kum) and semi-arid areas in Europe (e.g. the south-east of Spain). Figure 3.3, right map shows the added contribution of sea salt and wind-blown dust to the total PM_{10} mass. The contribution of these natural particles to PM_{10} is around 5-25% in central Europe, parts of Scandinavia off the sea and north-western Russia. The contribution increases to 20-30% in southern and eastern Europe In general, the importance of natural aerosols increases (to 50-75% and more) along the sea coasts due to the contribution of sea salt and in the south of Europe and southern parts of Ukraine and Russia, Caucasus and Kazakhstan due to wind-blown dust.

The middle map in Figure 3.3 shows that fine particles ($PM_{2.5}$) dominate PM_{10} mass practically all over Europe. The concentrations coarse particles are mostly a factor of 2 to 4 smaller than fine PM concentrations. The concentrations of coarse particles approach that of fine particles in the areas heavily affected by sea salt and wind-blown dust due to a considerable contribution of the natural coarse particles.

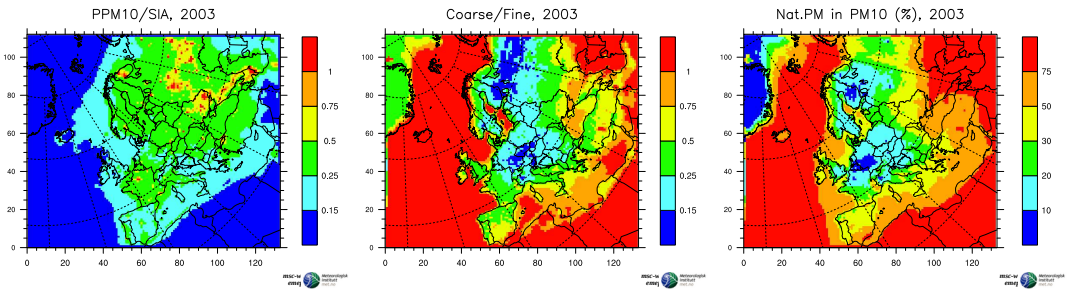

Figure 3.3: Ratios of calculated concentrations of primary PM_{10} to SIA (right panel) and coarse PM to fine PM (middle panel); and contribution of natural particles to PM_{10} (%) (right panel) in 2003.

3.2 European aerosol concentrations in 2003 compared to 2002

The EMEP model calculates annual mean concentrations of $PM_{2.5}$ and PM_{10} being 5-30% higher in 2003 than in 2002 almost all over of the EMEP area, reaching 30-35% (Figure 3.4, upper left map). The largest differences in $PM_{2.5}$ and PM_{10} concentrations (around 40%) are found on the Italian-French border. If only anthropogenic sources are considered, modelled $PM_{2.5}$ and PM_{10} concentrations are higher in 2003 than in 2002 in most parts of Europe (Figure 3.4, upper right map). The only areas, where calculated $PM_{2.5}$ and PM_{10} are somewhat lower in 2003 compared to 2002, are Spain, Ukraine, southern Russia, Kazakhstan, the Caucasus countries and Turkey.

The differences in air concentrations of aerosols arise from a combined effect of emission changes and meteorological variability. The changes in national emissions typically drive the changes in concentrations of anthropogenic particles. The production of natural particles, namely sea salt and wind-blown dust, is governed by the surface wind speed. Atmospheric dynamics (wind advection and turbulent mixing) determines pollutants dispersion and transport. Among the meteorological parameters affecting particle air concentrations are precipitation amount and surface stress (momentum flux), which determine respectively particle wet and dry removal (with wet deposition being the most important removal process for particles). Also the ambient temperature and relative humidity affect PM concentrations as they control the formation of semi-volatile ammonium nitrate (more NO_3^- and NH_4^+ aerosols are formed at lower temperature and higher relative humidity).

3.2.1 Effect of emission changes between 2002 and 2003

As seen from Figure 3.4, middle panels, both SIA and primary PM contribute to the enhancement of calculated anthropogenic $PM_{2.5}$ and PM_{10} concentrations in 2003. Partly, these differences are due to emission changes between 2002 and 2003. Larger 2003 emissions of SO_x in Sweden, Finland and Denmark, emissions of NO_x in Nor-

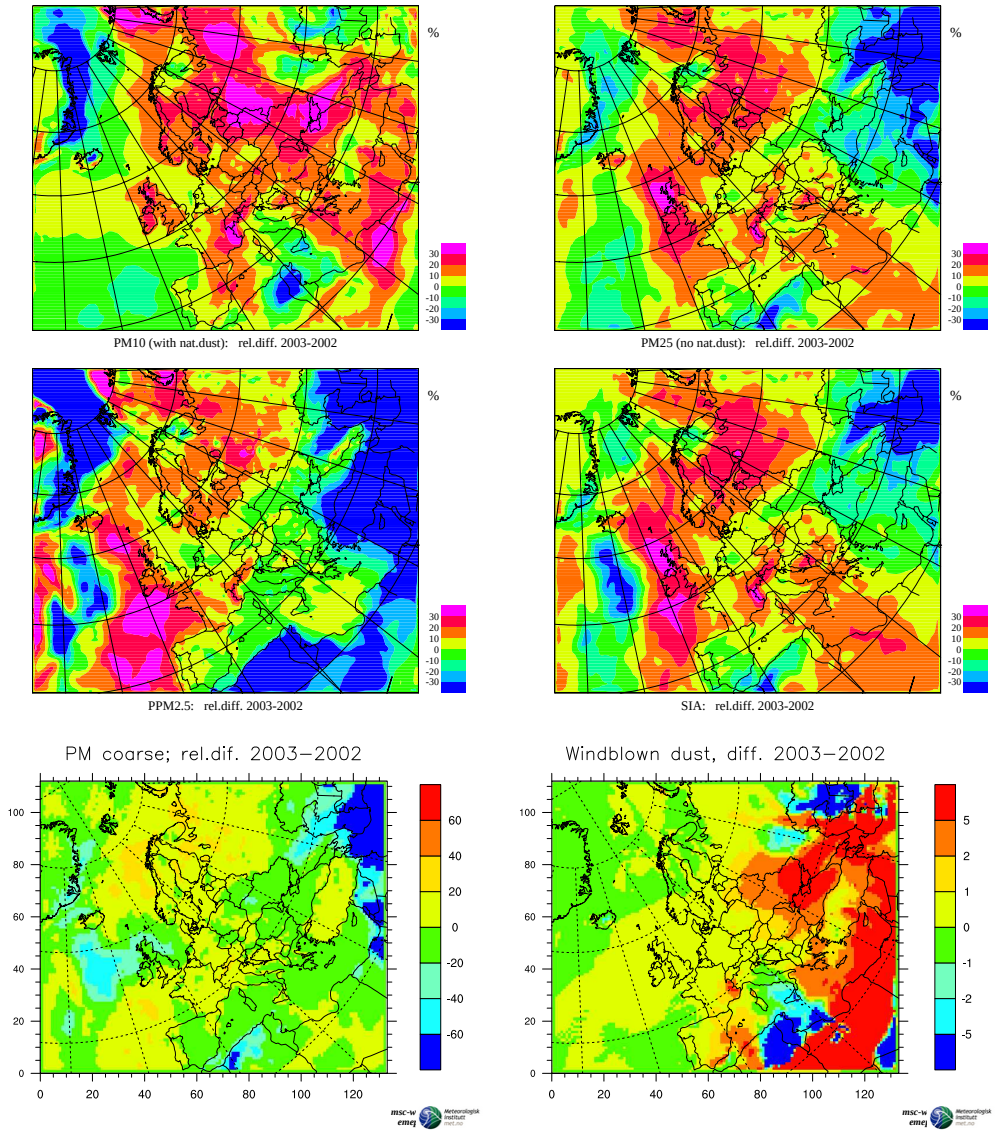

Figure 3.4: Differences between annual mean concentrations of: upper panels - PM_{10} and $PM_{2.5}$ (including wind-blown and Saharan dust), middle panels - primary $PM_{2.5}$ and SIA, and lower panels – coarse PM (units: %) and natural wind-blown and Saharan dust (units: μg/m^3). in 2003 and 2002. μ

way, Finland, Belarus, Latvia and Lithuania (Figure 3.6, upper maps) are responsible for the higher SIA concentrations in Scandinavia, northern and central Russia, the Baltic countries and Belarus (Figure 3.4, middle left map). The increase in SO_x emissions in Poland and Germany is counteracted by the decrease in NO_x and NH_3 and thus does not resulted in more SIA formation. The model calculates some increase in SIA concentrations in Austria, Czech Republic, Slovakia and Hungry due to larger emissions of SIA gaseous precursors in 2003 in these countries. In the rest of Europe (e.g. France, England, Italy), gas emissions somewhat decreased or remained unchanged

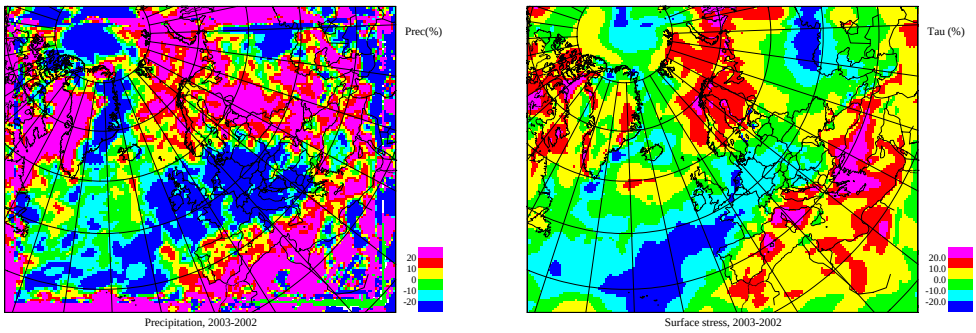

Figure 3.5: Relative differences between annual accumulated precipitation and mean momentum flux in 2003 and 2002. Units: %.

from 2002 to 2003. However, calculated SIA concentrations are larger in central and southern Europe in 2003 than in 2002, which is due to the particular meteorological conditions in 2003 (see 3.2.2).

Some increase in the emissions of primary $PM_{2.5}$ in Austria, Hungary, France, Sweden, Hellas and Moldova are reflected by the larger calculated $PPM_{2.5}$ concentrations in 2003 than 2002 in these areas (Figure 3.4, middle left map). The lower $PPM_{2.5}$ in Italy is a consequence of lower $PM_{2.5}$ emissions in the country in 2003. The emissions of coarse PM were larger in 2003 than in 2002 in Austria, Hungary, Germany, France, Portugal, Ireland and the Nordic countries (Figure 3.6, lower right map). This caused somewhat higher coarse PPM concentrations in those areas (Figure 3.4, lower right map). In Turkey, $PM_{2.5}$ emissions decreased, while coarse PM emissions increased in 2003 compared to 2002, resulting in lower $PPM_{2.5}$ and higher coarse PM concentrations in 2003 in Turkey.

3.2.2 Effect of meteorological variability between 2003 and 2003

Annual concentrations It is seen from Figure 3.4 that the overall distribution pattern of differences in $PPM_{2.5}$ concentrations resembles that of differences in SIA concentrations between 2003 and 2002. This can be attributed to the effect of meteorology on the pollutants air concentrations. Comparison of meteorological fields reveals that in central, western, southern and south-eastern Europe (except for Spain), less precipitation and more stable atmospheric conditions were in 2003 compared to 2002. According to model calculations, these conditions caused the larger concentrations of SIA, $PPM_{2.5}$, as well as total $PM_{2.5}$and PM_{10}, despite smaller emissions in several countries in 2003 than 2002 (as shown above). Inversely, in Spain, more precipitation and unstable atmosphere caused lower concentrations of all PM components in 2003.

On the other hand, a large area stretching over Scandinavia, the Baltic countries, Byelorussia, Ukraine and most of Russia experienced more precipitation and more unstable surface layer in 2003 than in 2002 (Figure 3.5). This suggests more efficient wet and dry removals of the pollutants, so that the effect of meteorology is to lower

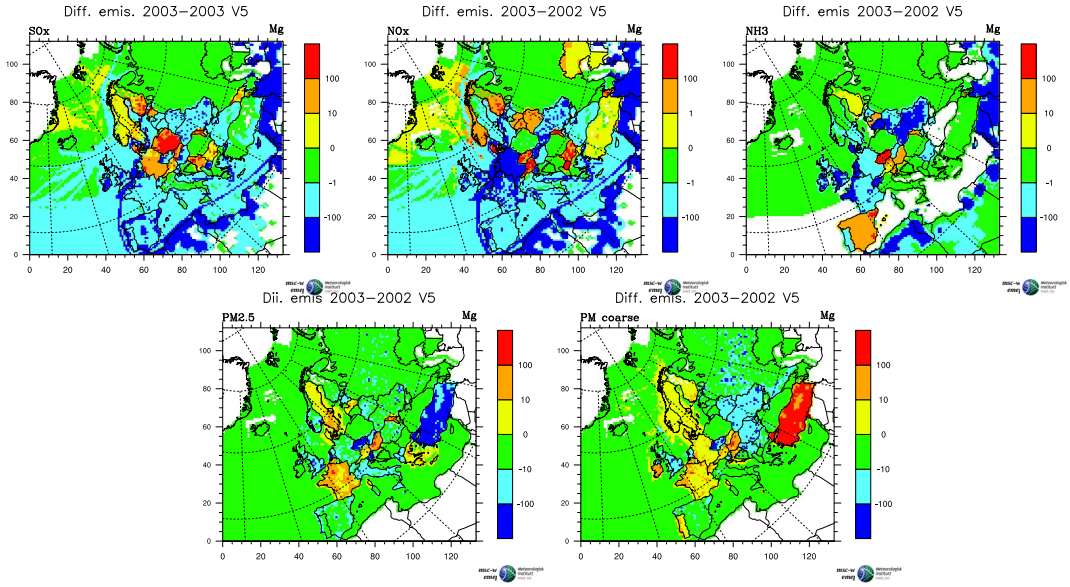

Figure 3.6: Differences in emissions of SO_x, NO_x, NH_3, primary $PM_{2.5}$ and coarse PM between 2003 and 2002. Units: tonnes.

particle air concentrations in 2003 compared to 2002 over this area. However, over Northern Europe, Baltic countries and northern Russia, calculated concentrations of the PM components are larger in 2003 than in 2002 (Figure 3.4, middle map). As we show below, this is probably due to the effect of high winter concentrations under specific meteorological conditions in 2003.

The emissions of dust particles from deserts and agricultural fields strongly depend on meteorological conditions. The mobilisation of soil particle is driven by wind and it is supressed by soil moisture, which depends on the precipitation. Overall, the mean average concentrations of wind-blown mineral dust are higher in 2003 than in 2002 (Figure 3.4, lower right map). Much of this dust is emitted in the northern Africa. Also, more wind-blown dust is produced in the semi-arid areas around French-Italian border and in south-eastern Spain due to particularly dry summer conditions in 2003.

Monthly concentrations Since grid distribution and sector allocation of emissions are the same for both years, increase/decrease in national emissions should affect air concentrations in the same manner through the whole year. However, due to meteorological variability the differences in seasonal/monthly concentrations do not necessarily follow the emission changes. Indeed, the differences between monthly mean concentrations fields are found to be considerably larger than between annual mean fields in 2003 and 2002

This is exemplified in Figure 3.7 for $PPM_{2.5}$ and SIA for the months of January, February and March. Figure 3.7 (two lower rows) presents a series of maps with relative differences between monthly mean concentrations of SIA and $PPM_{2.5}$ in 2003 and

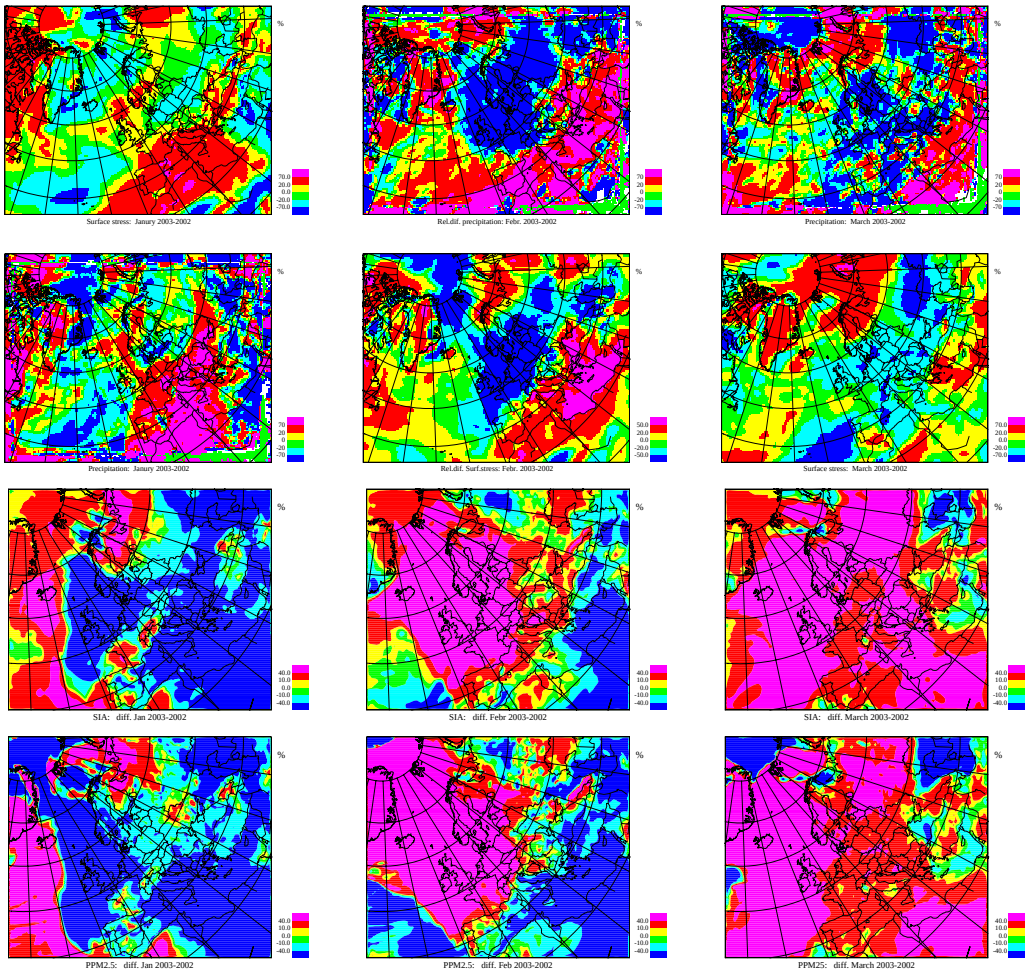

Figure 3.7: Relative differences in precipitation (upper panel), momentum flux (second upper row), SIA (third row) and primary $PM_{2.5}$ (lower panel) in January, February and March between 2003 and 2002. Units: %.

2002. Differences are also shown in monthly mean fields of precipitation amount and surface stress between those years (Figure 3.7, two upper rows). More precipitation is a major reason for lower SIA and $PPM_{2.5}$ concentrations in January 2003 than in January 2002 almost all over Europe, but the north of Scandinavia and Russia (Figure 3.7, left panels). In several areas, also less stable atmospheric conditions and thus more efficient dry deposition contributes to the lower SIA and $PPM_{2.5}$ in January 2003. Conversely, modelled concentrations of SIA and $PPM_{2.5}$ are much higher in February 2003 compared to February 2002 in central and northern Europe and north-western Russia as there was less precipitation and more stable surface layer restraining wet and dry particle removal (Figure 3.7, middle panels). It is worth pointing out that it is largely due to the effect of these high PM concentrations in February-March, that annual mean PM concentrations are higher in 2003 than in 2002 in these areas (as

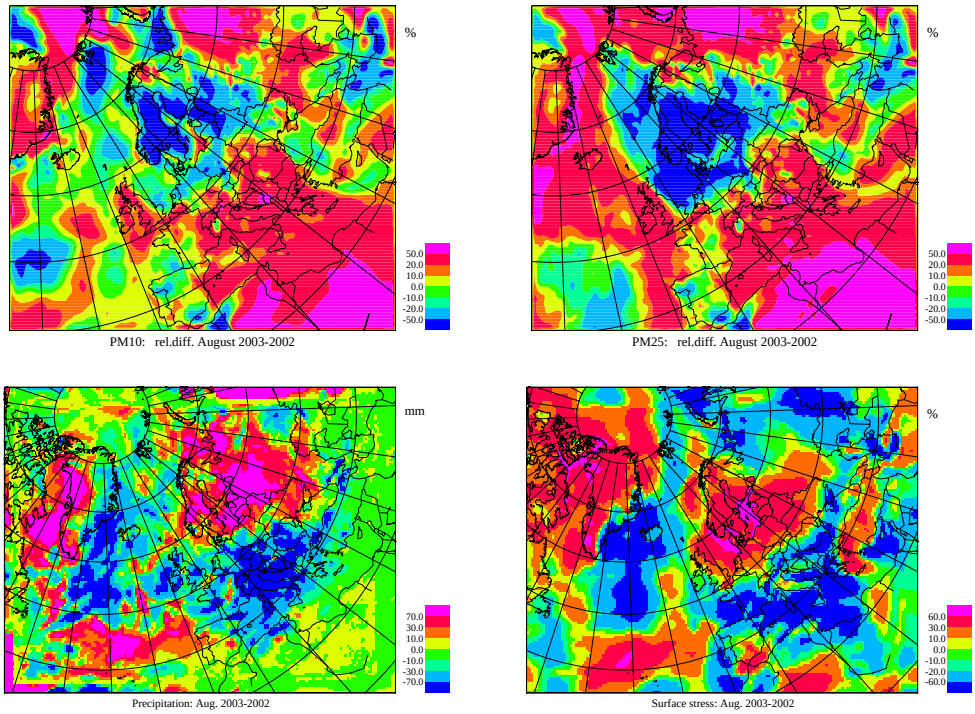

Figure 3.8: Relative differences in the concentrations of PM_{10} and $PM_{2.5}$ (upper panels) and in precipitation amount and momentum flux (lower panels) in August between 2003 and 2002. Units: %.

shown in the previous section). In Mediterranean countries, SIA and $PPM_{2.5}$ is lower in due to more intensive wet and dry deposition in February 2003 than in 2002. These dry and stable conditions persist also in March 2003, influencing now practically all Europe. As expected, modelled SIA and $PPM_{2.5}$ concentrations are higher in March in 2003 than in 2002 (Figure 3.7, right panels). Additionally, lower surface temperatures (see Figure 2.9), leading to more NO_3^- and NH_4^+ formation, contribute to enhanced SIA concentrations in February and March in 2003 compared to 2002.

August 2003 was characterised by exceptionally hot and dry weather conditions in central, southern and western Europe, and also in Russian Federation eastward from Moscow, while it was relatively cool and rainy in Scandinavia, Poland, the Baltic countries, Byelorussia and Ukraina (Figure 2.9). The strongest heat hit France, Switzerland and northern Italy and Spain, where the month mean temperature were up to 4-5 $^\circ$C higher in August 2003 than in 2002. The driest conditions occur in Italy and Balkan countries. The combination of little precipitation (see Figure 3.8, lower left map) and stable atmospheric conditions (Figure 3.8, lower right map) resulted in higher levels of $PM_{2.5}$ and PM_{10} in 2003 compared to 2002 in Mediterranean countries, Balkan countries, parts of France and Spain (Figure 3.8, upper maps).

Conversely, due to the rainy weather and unstable atmosphere $PM_{2.5}$ and PM_{10} concentrations in August 2003 were lower than in 2002 in the Nothern Europe, Baltic

countries, Byelorussia and north-west of Russia.

A considerable inter-month variability is found for the differences in wind-blown dust concentrations between 2003 and 2002. Much larger emissions of wind-blown dust in the semi-arid regions of Spain and south-eastern France take place in August in 2003 than in 2002. In March and August, the transport of mineral dust from northern Africa to Europe is less important, while in December it is larger in 2003 than in 2002. It is interesting to see different transport patterns of the wind-blown dust from the Kara Kum and Kyzyl Kum deserts, depending on the mean monthly winds in March and August.

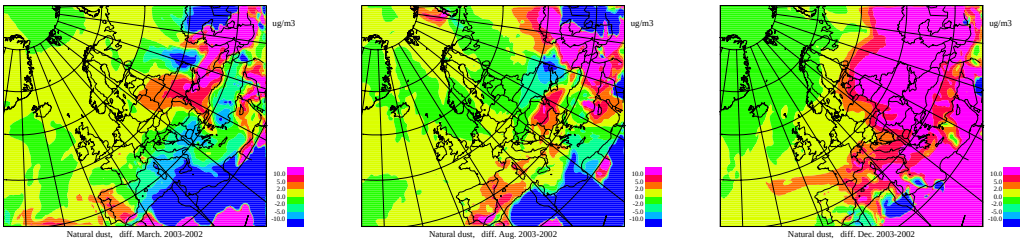

Figure 3.9: Difference in the concentrations of wind-blown dust between 2003 and 2002 in March, August and December. Units: μg/m^3.

3.3 Evaluation of the model performance for 2003

In this section we present the comparison of model results for particle concentrations with observations in 2003. We also compare the model performance for 2003 with its performance for 2002 and 2001. The year of 2003 is characterized as an extreme year with respect to the past European climatology in some areas (see Chapter 5). As it was said above, large parts of central, western and southern Europe were hit by a heat wave, which brought about a very hot and dry weather in July-August 2003. Here, we show how the model manages to reproduce aerosol concentrations in the exceptional weather conditions.

3.3.1 Annual mean scatter plots

The scatter-plots of model calculated annual mean concentrations of PM_{10}, $PM_{2.5}$ and SIA versus EMEP measurements in 2003 are shown in Figure 3.10 (right side plots). The modelled PM_{10} and $PM_{2.5}$ include SIA (SO_4^{2-}, NO_3^- and NH_4^+), primary anthropogenic PM, sea salt, wind-blown mineral dust from deserts and agricultural soils and particle water.

The model underestimates observed PM_{10} concentrations by 24% and $PM_{2.5}$ concentrations by 15%. In fact, these biases are lower than those for 2002, which are -26 and -19% respectively. The spatial correlation coefficients between modelled and

measured PM_{10} and $PM_{2.5}$ are as high as 0.73 and 0.80 in 2003, compared to the biases of 0.70 and 0.78 in 2002.

The improvement in model representation of regional PM concentrations is primarily due to accounting for wind-blown dust in the model. Particularly, the model calculates better PM concentrations at Spanish sites, where natural dust is an important PM element. This considerably improves the general model performance because Spanish sites are overrepresented among the EMEP sites with PM measurements. The remaining model underestimation of PM_{10} and $PM_{2.5}$ is largely because secondary organic aerosols (SOA) have not yet been included in the EMEP Unified model. Also primary biogenic organic aerosols, such as pollen, spores and fungi, which can be significant contributors to PM_{10} mass, are not accounted for.

The following should be pointed out. The model performance substantially improves with respect to PM_{10} and $PM_{2.5}$when wind-blown dust and particle water are accounted for. This indicates that the model gives a reasonable estimate of wind-blown dust concentrations and particle water. However, the proper validation of model results for wind-blown dust and particle water with measurements is not feasible at present because of the lack of measurements. Therefore there are still uncertainties related to the model calculations of wind-blown mineral dust and particle water.

SIA is by far the most important component of background PM. The scatterplots for SIA compare annual mean calculated concentrations with observations the at EMEP stations where inorganic aerosols SO_4^{2-}, NO_3^- and NH_4^+ were measured concurrently in 2003 (Figure 3.10, lower plots). In 2003, the model overestimates measured SIA by 22%, which is somewhat greater than overestimation of observations in 2002 (16%). The spatial correlation between modelled and observed SIA is 0.87 in 2002 and 0.89 in 2003.

Another important component of PM is primary particles from anthropogenic sources. Elemental carbon is a good candidate component for validation of primary PM emissions, however it is presently unfeasible to verify primary anthropogenic OC. The model calculated EC and OC concentrations are compared with measurements from NILU's OC/EC campaign from July- 2002 through July 2003 (Yttri, 2004 and 2005). The measurements were taken one day a week. For 2003, only stations with at least 9 days with measurements were included in the scatter-plots. Calculated EC and OC show quite good spatial correlation with measured concentrations (Figure 3.11), which indicates a reasonably good description of the main sources of EC/OC emissions. The better correlation for EC in 2002 than in 2003 is probably due to less significant statistics in 2003. EC is underestimated by the model by 46% in 2002 and by 30% in 2003. The model's negative bias for OC is much larger: -87 and -83% in 2002 and 2003. Notice that emissions of carbonaceous particles from wild forest fires are not accounted for. Furthermore, the calculated OC concentrations are much lower than measured concentrations because they do not include primary coarse OC from anthropogenic sources, secondary organic aerosols (SOA) and primary biogenic OC.

The yearly and seasonal statistics of model performance for PM_{10}, $PM_{2.5}$ and SIA concentrations in the years 2001, 2002 and 2003 are summarised in Table 3.1. It can

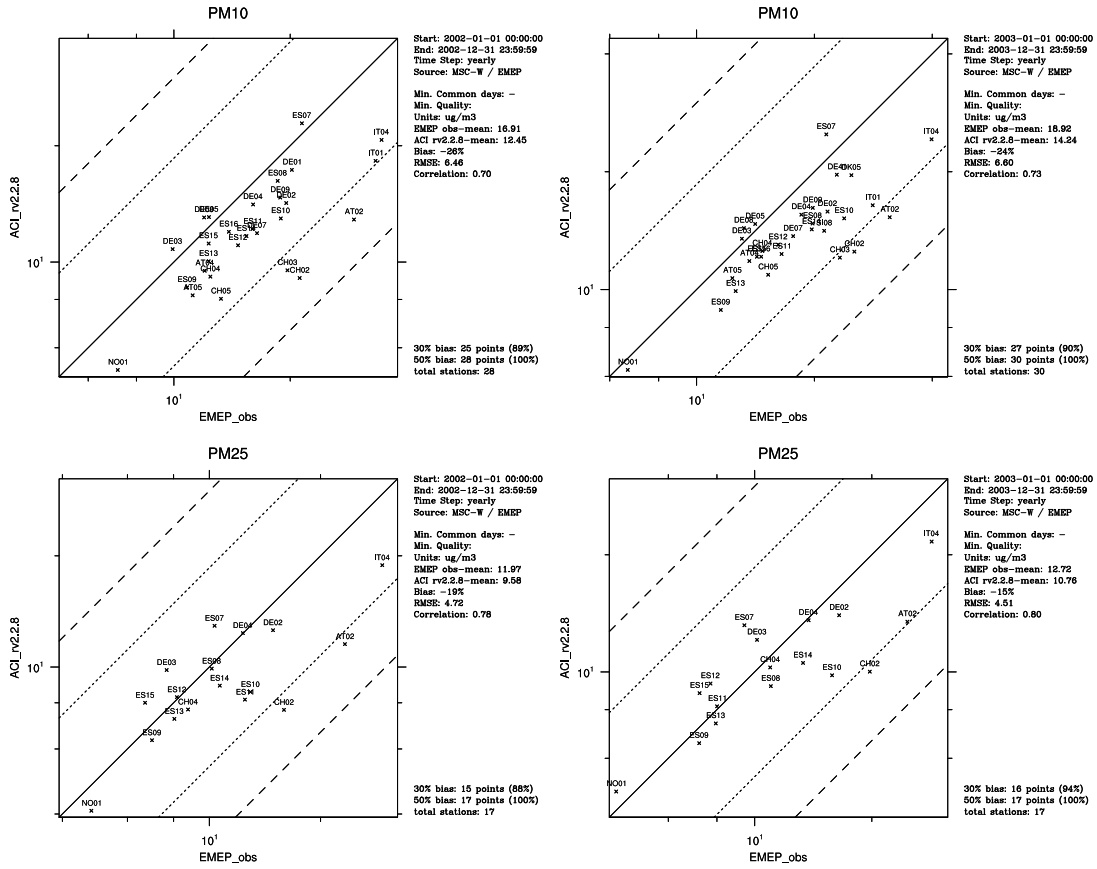

Figure 3.10: Scatter-plots of calculated vs. measured PM_{10}, $PM_{2.5}$ and SIA for EMEP sites in 2002 and 2003. Units: μg/m^3.

be seen that the model performance is quite stable for these years and seasons. Model negative bias for PM is largest in summer; it is around -40% for PM_{10} and around -28% for $PM_{2.5}$.The spatial correlation coefficient between calculated and measured PM_{10} and $PM_{2.5}$ varies mostly between 0.41 and 0.68 in different seasons. Rather robust statistics indicate that the model manages fairly well to account for the effect of inter-annual meteorological variability on PM concentrations.

As far as the model performance in summer 2003 concerned, model underestimation of PM_{10} and $PM_{2.5}$ is only insignificantly larger in 2003 then in 2002 and 2001. The spatial correlation for PM_{10} and $PM_{2.5}$ is only slightly lower in summer 2003 compared to 2002 and 2001. The model results for SIA compares slightly better with observations in summer 2003 than in 2002 and 2001.

3.3.2 Monthly variation of aerosol concentrations

Monthly time-series of modelled and measured PM_{10} and $PM_{2.5}$, averaged over the EMEP sites with PM measurements in 2002 and 2003, are presented in Figure 3.12.

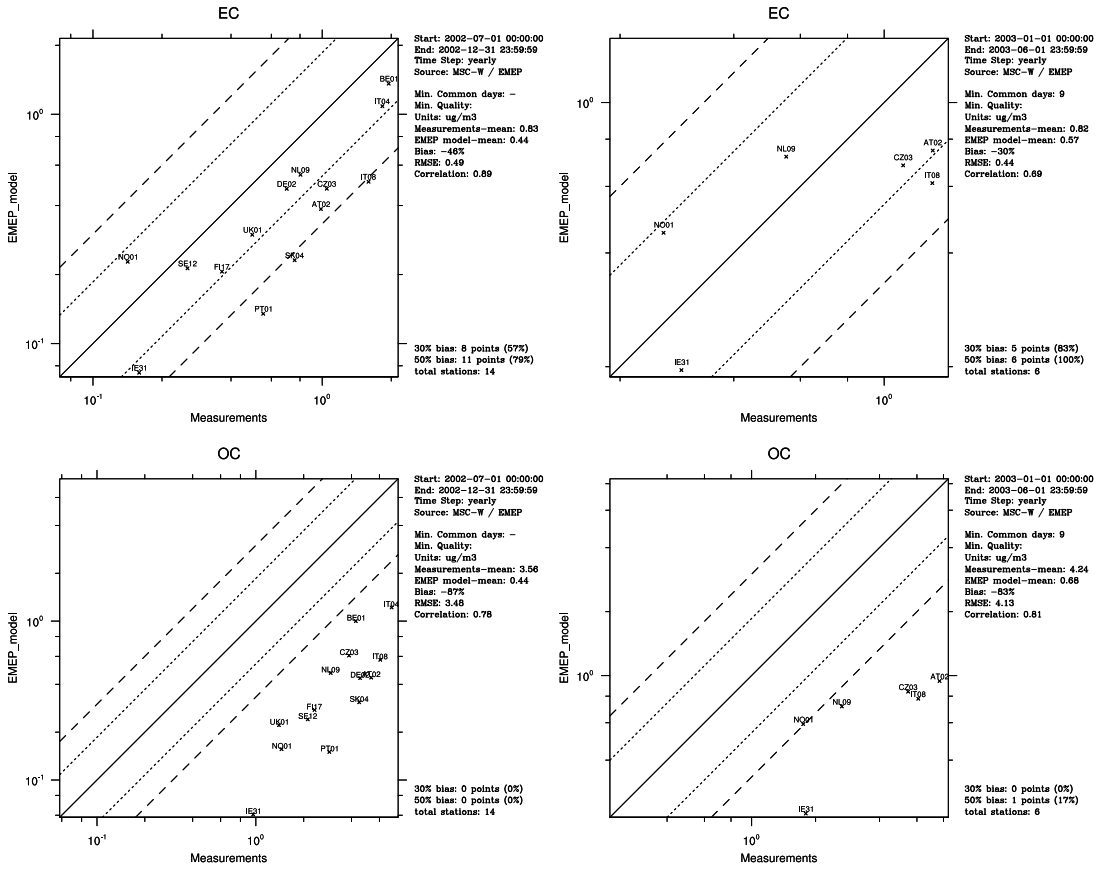

Figure 3.11: Scatter-plots of calculated vs. measured EC and OC in July-December 2002 and January-April 2003 (measurements are 24-hour averages taken one day a week). Units: μg(C)/m^3.

The three curves for calculated PM concentrations represent: dry PM without wind-blown dust (magenta), dry PM including wind-blown dust (black), and PM concentrations including wind-blown dust and particle water (blue). Both calculated and observed summer concentrations of PM_{10} and $PM_{2.5}$ are somewhat higher in 2003 than in 2002. In this data set, this is mostly due to higher PM concentrations at Swiss and several Spanish stations, which experienced the most hot and dry weather in July-August 2003. At these sites, observed daily PM_{10} concentrations were 3-4 μg/m^3 higher and $PM_{2.5}$ concentrations were 2-3 μg/m^3 higher in July-August in 2003 than the correspondent values in 2002.

The model manages quite well to describe the observed monthly variation of PM_{10} and $PM_{2.5}$ in both of the years. The model tends to underestimate PM_{10} and $PM_{2.5}$ concentrations in the warm period, from April/May to August. In the cold season, calculated PM_{10} and $PM_{2.5}$ concentrations are quite close to the measured values, which is partly resulting from model overestimation of SIA (Figure 3.13). Too high calcu-

Table 3.1: Yearly and seasonal statistics of the model performance

PM10	2003 (30 sites)			2002 (28 sites)			2001 (26 sites)		
Period	Obs	Bias	Corr	Obs	Bias	Corr	Obs	Bias	Corr
Yearly	18.92	-24	0.73	16.91	-26	0.70	16.94	-24	0.67
Daily	18.89	-24	0.55	16.85	-26	0.52	16.5	-24	0.5
JanFeb	20.09	-17	0.57	16.46	-18	0.41	18.36	-18	0.53
Spring	21.51	-24	0.65	18.64	-25	0.61	14.51	-22	0.44
Summer	20.14	-40	0.54	18.76	-39	0.56	17.93	-37	0.61
Autumn	15.97	-13	0.49	13.83	-17	0.53	16.68	-16	0.55
PM25	2003 (18 sites)			2002 (317sites)			2001 (17 sites)		
Period	Obs	Bias	Corr	Obs	Bias	Corr	Obs	Bias	Corr
Yearly	12.54	-15	0.81	11.97	-19	0.78	12.12	-21	0.82
Daily	12.42	-14	0.56	11.7	-18	0.53	11.86	-20	0.5
JanFeb	13.67	-10	0.63	12.24	-18	0.43	16.84	-26	0.56
Spring	13.28	-11	0.68	12.55	-17	0.66	10.07	-20	0.55
Summer	13.64	-29	0.41	12.87	-25	0.44	12.1	-27	0.53
Autumn	10.66	-3	0.49	9.4	-12	0.52	11.63	-12	0.47
SIA	2003 (18 sites)			2002 (18 sites)			2001 (20 sites)		
Period	Obs	Bias	Corr	Obs	Bias	Corr	Obs	Bias	Corr
Yearly	4.22	22	0.87	3.99	16	0.89	4.42	8	0.91
Daily	4.29	21	0.67	3.97	16	0.69	4.27	8	0.66
JanFeb	5.77	40	0.58	4.45	37	0.70	4.59	40	0.61
Spring	5.08	21	0.64	4.32	19	0.74	4.39	5	0.7
Summer	3.47	-4	0.75	3.58	-3	0.64	3.97	-6	0.73
Autumn	3.67	22	0.78	3.29	17	0.70	4.17	6	0.68

lated PM_{10} and $PM_{2.5}$ concentrations in November can be due to model overestimation of SIA and also wind-blown dust.

It can be seen from Figure 3.13 that the model over-prediction of SIA in cold period is due to too high calculated concentrations of NH_4^+, and especially NO_3^-. The model calculations agree well with measurements that the highest NO_3^- and NH_4^+ concentrations occur in February and March in 2003. Comparison of meteorological fields reveals that it was much less precipitation and more stable atmosphere in central, northern and north-eastern Europe, where most of the sites with SIA measurements are located, in February-March 2003 compared to 2002 (see the previous section). This means less efficient wet and dry scavenging of aerosols and therefore larger air concentrations. Also, those areas experienced a lower surface layer temperature favouring a larger aerosol formation in February-March 2003 compared to 2002. However, the equilibrium between gas and aerosol phase appears to be too sensitive to the temperature changes, producing too many particles in model.

In summary, the analyses of model validation for 2002 and 2003 has shown that the model is quite capable of reproducing observed PM_{10} and $PM_{2.5}$ concentrations under

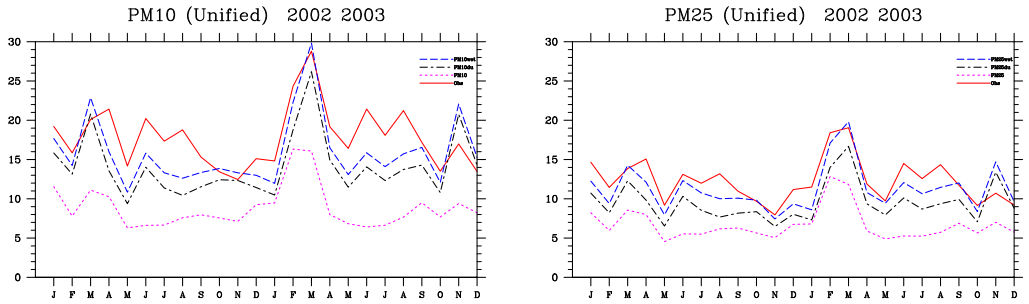

Figure 3.12: Monthly time-series of calculated and measured PM_{10} and $PM_{2.5}$ in2002 and2003. Here, calculated PM: magenta dots - dry PM without wind-blown dust, black dot-dashed line - dry PM including wind-blown dust, and blue dashed line - PM including wind-blown dust and particle water; red solid line – measurements (for the number of stations with measurements in different years see Table 3.1). Units: μg/m^3.

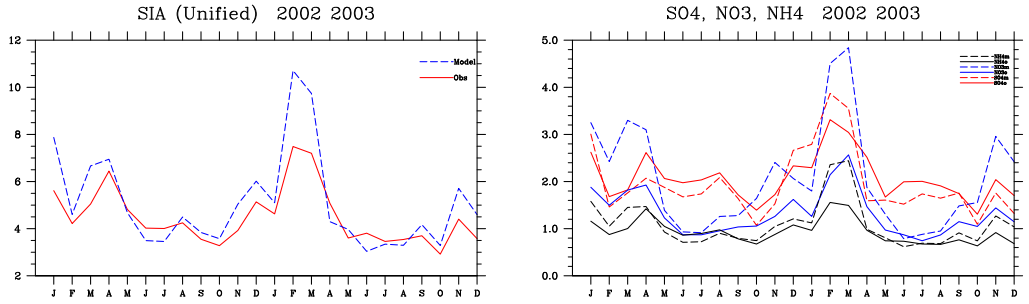

Figure 3.13: Monthly time- of calculated (dashed lines) and measured (solid lines) SIA and SO_4^{2-} (red), NO_3^- (blue) and NH_4^+ (black) in 2002 and 2003. SIA is averaged over 18 sites, SO_4^{2-} over 74 and 72 sites in 2002 and 2003, NO_3^- over 25 and 35 sites and NH_4^+ over 20 sites. Units: μg/m^3.

variable meteorological conditions. The largest discrepancies between model calculations and measurements are found for NO_3^- and NH_4^+ aerosols, which are overestimated by the model in the cold period. Also, there are still uncertainties in calculations of EC and especially OC related to the uncertainties in anthropogenic PM emissions and missing emission sources of carbonaceous aerosols in the model.

3.3.3 Model calculations of PM under the heat wave conditions in 2003

To see how the model manages to reproduce PM concentrations in the hot and dry conditions of summer 2003 we compare daily time-series for PM_{10} and $PM_{2.5}$ at two Swiss sites and one Italian site in July-August 2002 and 2003 (Figure 3.14). For the whole July-August period, model bias for PM_{10} and $PM_{2.5}$ is roughly the same in 2003

and 2002 at these sites. At the Swiss sites, the model does calculate enhanced PM_{10} and $PM_{2.5}$ concentrations in 2003 compared to 2002. If we consider only the first half of August, when the weather was the most extreme, the model negative bias is just a little (2-8%) larger in 2003 than in 2002. At Ispra, the average calculated PM_{10} is somewhat lower in July-August in 2003 than in 2003. On the other hand, for the hottest 2 weeks in the first half of August the model calculates much PM_{10} in 2003 than in 2002. Still, the calculated PM_{10} concentrations are not as high as it was observed, and the underestimation of observations is about 10% larger in 2003 than in 2002.

The temporal correlation between calculated and measured $PM_{10}$and $PM_{2.5}$ is better in 2003 than in 2002 at the Swiss sites. At Ispra, the temporal correlation for dry PM_{10} is better, while it is lower for PM_{10} including particle water in 2003 compared to 2002. This suggests that further testing and verification of modelled particle water is needed. Also at the other EMEP stations, the performance of the model for PM_{10} and $PM_{2.5}$ is similar to or better in 2003 compared to 2002 (Tsyro (2004) and Tsyro (2005a)).

Concluding, for the most part the EMEP model is found to do a reasonable job calculating PM_{10} and $PM_{2.5}$ concentrations in the extreme conditions of summer 2003.

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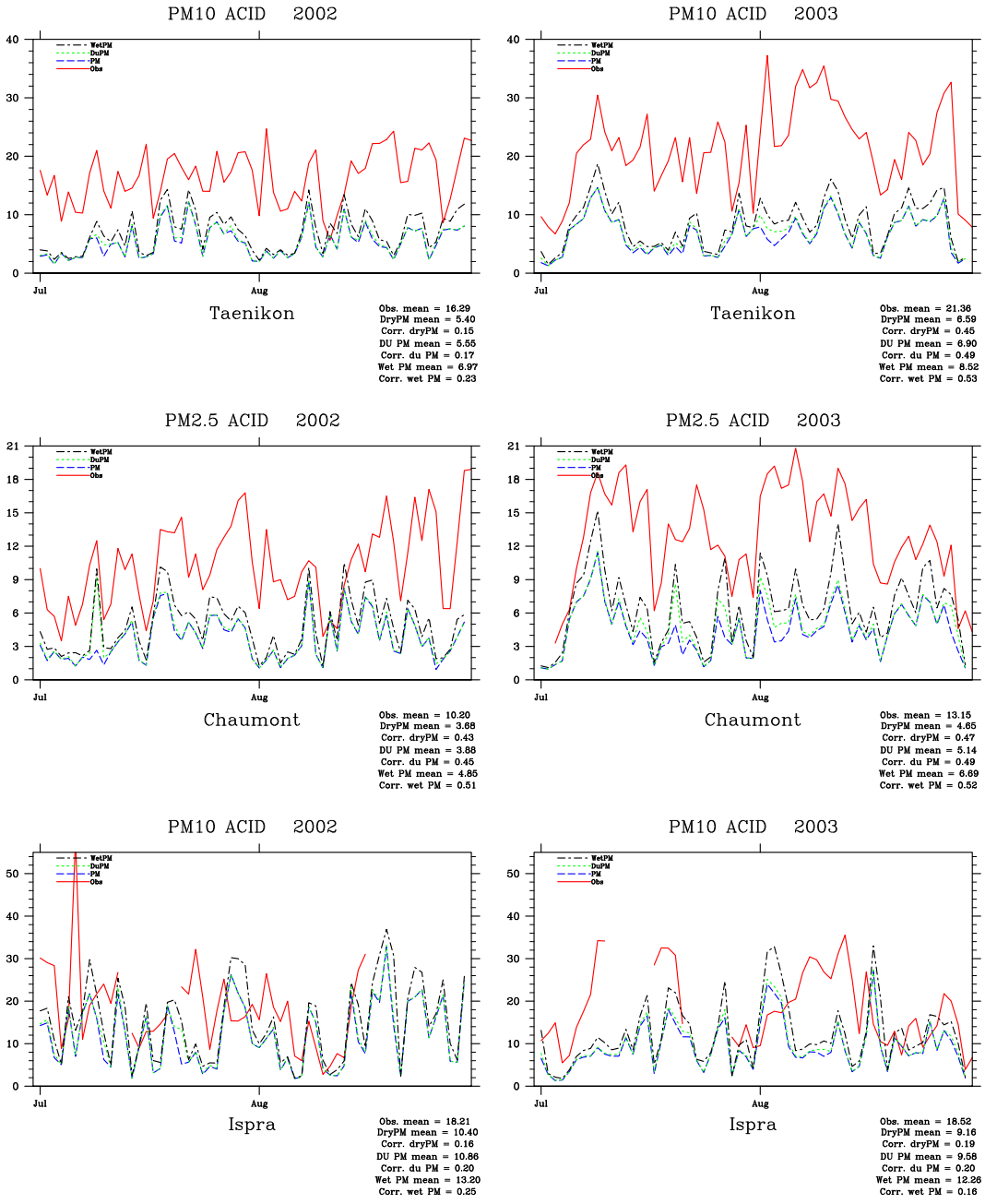

Figure 3.14: Daily time-series of PM_{10} at Taenikon (CH03) and Ispra (IT04) and $PM_{2.5}$ in Chaumont (CH04) in July-August in 2002 and 2003. Units: $\mu g/m^3$.

CHAPTER 4

Photo-oxidants, status in 2003

David Simpson, Jan Eiof Jonson and Hilde Fagerli

4.1 Introduction

This chapter examines the performance of the EMEP photo-oxidant model (revision rv2.3) for ozone and, to a limited extent, NO_2. The analysis focuses on comparisons against recent measurements (2002-3). We present results for two years for comparison purposes, because of the extreme nature of both meteorology and pollution concentrations in 2003. The summer of 2003 was exceptionally warm, in particular in central Europe, and this lead to much higher ozone than usual, especially in countries such as Switzerland (Fink et al. 2004, Solberg et al. 2005, Ordóñez et al. 2005). Unfortunately, commitments to other projects and CAFE has meant that we have not had time to analyse the EMEP model results for this summer yet. This work will be conducted though, since the summer of 2003 presents a good opportunity to test and understand model performance in extreme meteorological conditions.

Other work on photo-oxidants has been conducted in the framework of two EU projects which finalised in 2004. These projects are (a) CARBOSOL - focuses on carbonaceous aerosol over Europe. In this work the photo-oxidant model was linked up to a module for secondary organic aerosols, extending the previous work of Andersson-Sköld and Simpson (2001) and Simpson and Makar (2004) ; and (b) NOFRETETE - explored the links between nitrogen oxides emissions from forests, nitrogen deposition, and ozone formation.

Papers from NOFRETETE have been submitted for publication, and papers for CARBOSOL are in preparation at the time of writing. An additional paper on ozone trends has been accepted for publication Jonson et al. (2005).

4.2 Time-series for ozone

As in previous ozone model evaluations (Simpson, 1992, 1993, 1998, Simpson and Jonson, 1998, Jonson et al., 1998, 2002), we use the daily maximum ozone as the basis of our statistical evaluation. The reason for this is that the daily maximum usually represents the time when the boundary layer is well-mixed (mid-afternoon, typically), and so modelled and observed concentrations should be most comparable.

Table 4.1 presents a summary of the model results compared against observations for all sites within the EMEP network. The correlation coefficients between the daily maximum ozone values are presented. It should be noted that no attempt has been made in this table to screen out sites which are believed to be influenced by local sources or to suffer from poor data quality, except that comparisons are only shown for those sites which had more than 274 valid days of observations. These results will be discussed region by region below, with plots presented for selected sites.

Table 4.1: Comparison of Modelled Versus Observed Ozone for Year 2003. Concentrations are 12-monthly Means of Daily Maximum Ozone Values. Correlation coeffi cient (r) are also between daily max values. Only sites with more than 275 valid observation days (N) are shown.

Code	Station	N (days)	Obs. (ppb)	Mod. (ppb)	r
		*N*ordic Countries			
DK05	Keldsnor	364	39.45	39.37	0.83
FI09	Utoe	334	40.23	36.70	0.76
FI17	Virolahti II	355	37.67	35.61	0.69
FI22	Oulanka	352	37.87	32.89	0.62
FI37	Aehtaeri II	332	37.19	32.69	0.67
NO01	Birkenes	362	37.27	36.96	0.69
NO15	Tustervatn	328	41.80	36.03	0.62
NO39	Kaarvatn	333	41.09	37.19	0.36
NO41	Osen	353	37.22	35.36	0.61
NO42	Spitzbergen, Zeppelin	358	38.45	38.37	0.55
NO43	Prestebakke	361	39.54	37.20	0.75
NO55	Karasjok	364	40.65	34.99	0.52
NO56	Hurdal	362	37.41	34.24	0.67
SE11	Vavihill	358	40.85	37.55	0.74
SE12	Aspvreten	350	42.04	36.27	0.78
SE13	Esrange	364	40.17	34.93	0.49
SE32	Norra-Kvill	363	42.12	36.39	0.73

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Code	Station	N	Obs.	Mod.	r
SE35	Vindeln	358	39.77	33.13	0.60
	*E*astern European Countries				
CZ03	Kosetice	303	52.13	44.85	0.82
EE09	Lahemaa	354	39.14	33.35	0.59
EE11	Vilsandy	339	42.76	38.06	0.74
HU02	K-puszta	301	49.39	45.35	0.78
LT15	Preila	344	37.95	41.37	0.73
LV10	Rucava	337	34.89	37.56	0.56
PL02	Jarczew	364	42.18	39.45	0.82
PL03	Sniezka	364	50.54	40.79	0.74
PL04	Leba	364	42.87	40.42	0.79
PL05	Diabla Gora	356	40.56	37.24	0.78
RU18	Danki	350	31.59	36.32	0.63
SI08	Iskrba	344	50.99	45.27	0.80
SI31	Zarodnje	355	48.31	44.64	0.86
SI32	Krvavec	358	58.30	45.29	0.84
SI33	Kovk	345	49.24	45.70	0.82
SK04	Stara Lesna	350	47.54	41.25	0.69
SK06	Starina	360	49.04	41.11	0.78
SK07	Topolniky	361	50.74	44.23	0.82
	*C*entral and NW European Countries				
AT02	Illmitz	357	50.62	44.29	0.86
AT04	St. Koloman	357	51.75	44.41	0.81
AT05	Vorhegg	364	50.18	45.84	0.75
AT30	Pillersdorf	363	47.25	42.31	0.88
AT32	Sulzberg	351	54.02	45.05	0.79
AT33	Stolzalpe	347	49.81	45.05	0.75
BE01	Offagne	338	46.89	43.19	0.86
BE32	Eupen	341	47.05	41.52	0.87
BE35	Vezin	352	42.99	41.26	0.87
CH02	Payerne	364	47.37	45.33	0.81
CH03	Taenikon	364	47.04	44.93	0.80
CH04	Chaumont	363	54.97	45.34	0.83
CH05	Rigi	361	54.94	44.63	0.76
DE01	Westerland/Wenn.	363	45.79	36.66	0.71
DE02	Lang./Waldhof	363	44.47	38.91	0.89
DE03	Schauinsland	357	53.90	43.56	0.84
DE04	Deuselbach	364	46.86	43.16	0.89
DE07	Neuglobsow	355	42.44	39.07	0.87
DE08	Schmuecke	360	51.02	41.28	0.88
DE09	Zingst	363	40.74	33.89	0.65

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Code	Station	N	Obs.	Mod.	r
DE12	Bassum	363	39.25	39.45	0.87
DE26	Ueckermuende	358	41.00	38.88	0.82
DE35	Lueckendorf	358	46.39	40.95	0.86
DE39	Aukrug	346	38.98	37.79	0.85
FR08	Donon	358	52.02	43.60	0.85
FR09	Revin	362	44.68	42.58	0.88
FR10	Morvan	357	47.77	42.60	0.84
FR12	Iraty	311	55.04	43.82	0.77
FR13	Peyrusse Vieille	351	46.13	42.39	0.82
FR14	Montandon	355	44.65	44.54	0.79
GB02	Eskdalemuir	353	35.15	39.34	0.77
GB13	Yarner Wood	361	42.51	40.66	0.79
GB14	High Muffles	359	35.67	37.53	0.53
GB15	Strath Vaich Dam	315	43.98	38.70	0.74
GB31	Aston Hill	362	38.67	39.80	0.87
GB32	Bottesford	362	41.21	36.31	0.80
GB33	Bush	359	39.25	36.78	0.69
GB34	Glazebury	323	33.84	33.55	0.74
GB35	Great Dun Fell	361	40.44	39.13	0.65
GB36	Harwell	350	44.08	37.62	0.84
GB37	Ladybower	357	38.78	35.40	0.76
GB38	Lullington Heath	349	43.66	40.13	0.83
GB39	Sibton	333	40.42	38.27	0.77
GB43	Narberth	318	35.14	41.26	0.74
GB44	Somerton	354	42.28	39.09	0.84
GB45	Wicken Fen	361	41.09	37.46	0.81
IE31	Mace Head	351	43.26	41.16	0.73
NL09	Kollumerwaard	360	35.88	38.69	0.86
NL10	Vreedepeel	360	37.20	38.70	0.89
	*M*editerranean Countries and Portugal				
ES07	Viznar	352	46.35	46.02	0.70
ES08	Niembro	339	40.58	46.84	0.36
ES09	Campisabalos	348	48.33	45.64	0.77
ES10	Cabo de Creus	347	55.73	46.35	0.56
ES11	Barcarrota	358	45.63	44.73	0.85
ES12	Zarra	356	49.92	46.73	0.76
ES13	Penausende	351	51.68	44.43	0.83
ES14	Els Torms	355	51.03	47.68	0.84
ES15	Risco Llano	354	53.00	45.19	0.83
IT01	Montelibretti	355	54.44	50.10	0.84
PT04	Monte Velho	317	50.74	45.51	0.52

4.2.1 Nordic Sites

Figure 4.1 presents time-series plots of modelled versus observed daily maximum ozone concentrations for a number of Nordic sites for the years 2002 and 2003. Figures 4.2 presents plots for 2003 for some further sites. In general, the model reproduces the observed concentrations rather well at these Nordic sites, and in some cases the agreement is excellent. A general feature is that the correlation coefficients are better for 2003 than 2002, but the model has a stronger tendency to underpredict in 2003 than in 2002. Most of this underprediction occurs in the Spring-time though. For example. the site Tustervatn (NO15), which is located at around 66°N, is usually predicted very well by the EMEP model, and this has suggested in the past that the procedures used for setting boundary conditions in the model (making use of observations from Mace Head, at only 53°N, see Simpson et al., 2003) perform well even in the far northern part of the domain. This good prediction was seen in 2002 (not shown). However, in 2003, there are clear problems from Feb-April, suggesting that the model's boundary conditions may be too low for Northern sites int his period. An ozone episode of ca. 80 ppb is also seen at several sites in April 2003. This episode is seen at sites further south also, although not so much at Mace Head. The reasons for this episode will be explored in future work.

4.2.2 Eastern European Sites

Figures 4.3-4.4 present time-series plots of modelled versus observed daily maximum ozone concentrations for a number of Eastern European sites for the year 2003.

The model is able to reproduce the time-series for most of these sites rather well, with correlations for all Eastern European sites ranging from r =0.56 (LV10) to r =0.86 (SI08). Performance is poor for only two of the sites shown, Lahemaa (EE09) in Estonia and Danki (RU18) in Russia. For site LV10 the very low observed values seen in May look very unusual, and are likely a sign of measurement problems. In general it is not clear why some of these sites are poorly reproduced when compared to other sites in the Baltic region (including the Finnish site Utö). The Russian site, RU18, lies close to Moscow and here the agreement is quite good, except for a large model over-prediction in July. Results for sites in SE Europe are generally good, be a tendency for the model to show lower ozone values than the observations, especially in the spring-time period which was problematic for the Northern sites.

4.2.3 Central and North-West European Sites

Figure 4.5 presents time-series plots of modelled versus observed daily maximum ozone concentrations for a number of central and North-West European sites for the years 2002 and 2003. Figure 4.6 presents similar plots for some further sites for 2003.

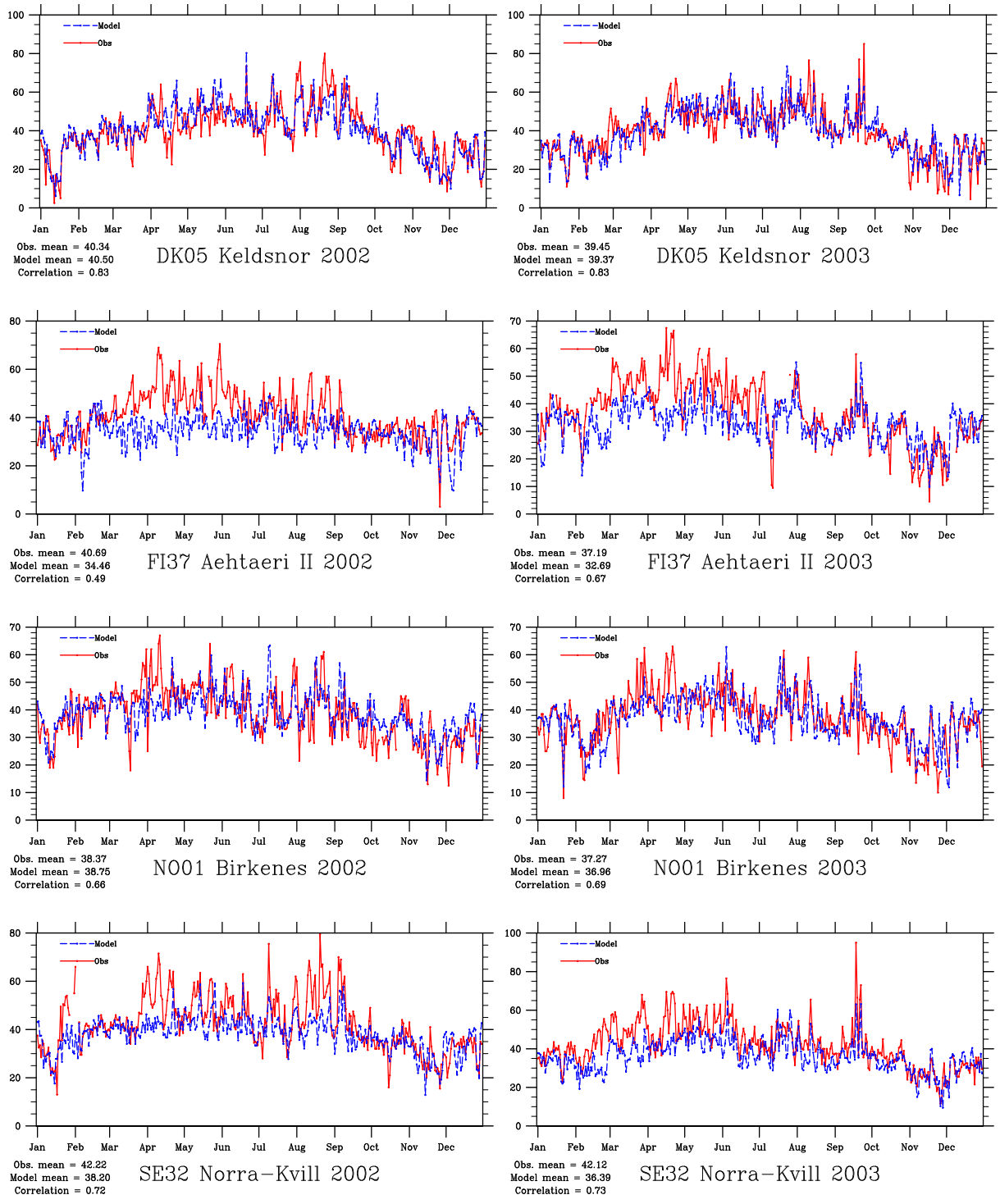

Figure 4.1: Modelled versus Observed Daily Max Ozone (ppb), Nordic Sites, 2002 and 2003

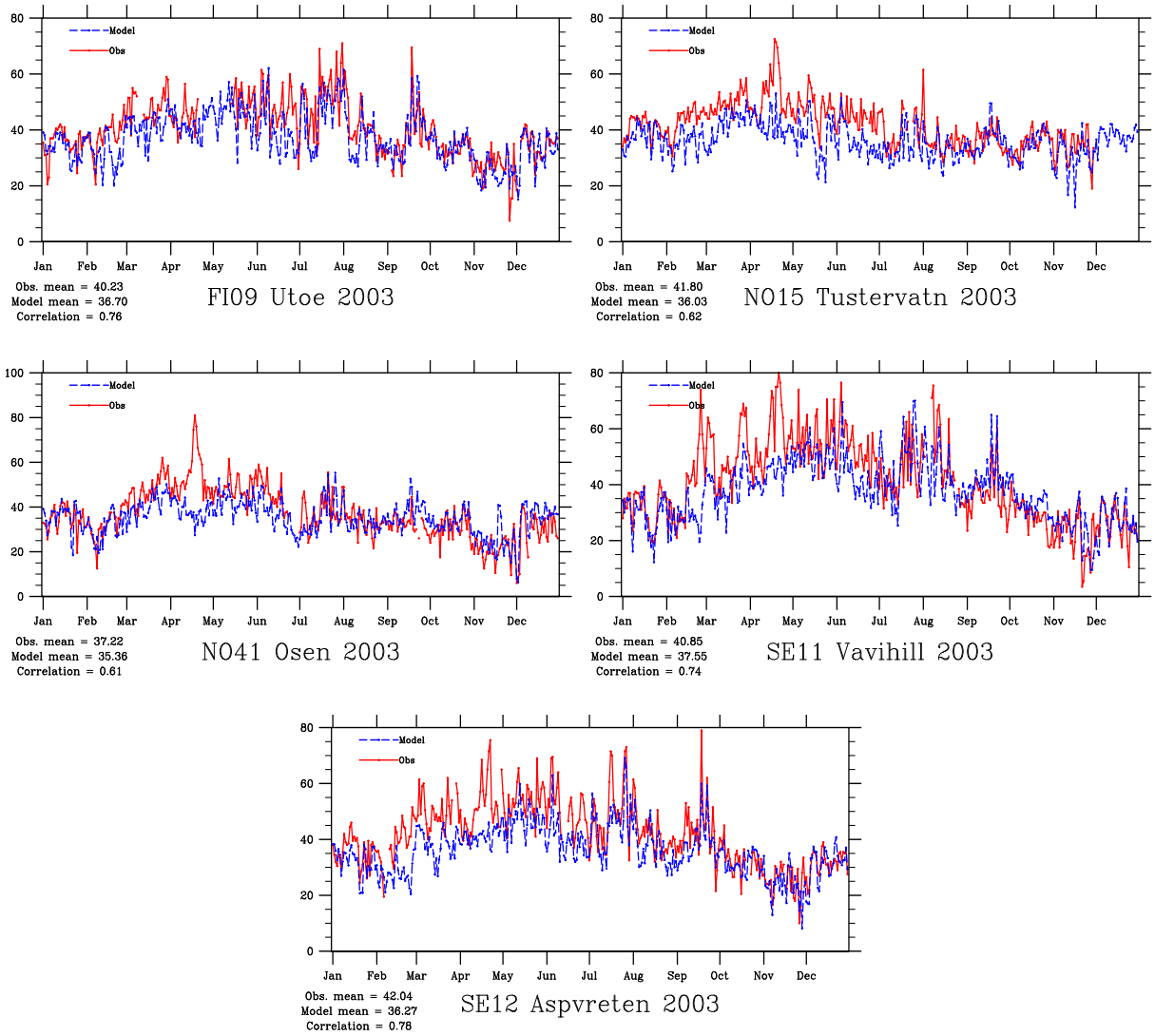

Figure 4.2: Modelled versus Observed Daily Max Ozone (ppb), Nordic Sites, 2003

The model performs well for all of these sites for 2002, with correlation coefficients often exceeding 0.8 and with good reproduction of seasonal cycles. There seems to be some tendency to underpredict the highest ozone episodes in 2002, but the occurrence of most episodes is reproduced well. In 2003 the correlation coefficients actually increase at most sites, but there is a systematic underestimated of observed ozone at many sites, especially in the south. For example, the mean ozone at Illmitz (AT02) is predicted within ca. 1ppb in 2002, but underpredicted by over 5 ppb in 2003.

Sites further north often show very good agreement, even for 2003. For example, Aston Hill in the UK has r =0.87, mean ozone within 1 ppb for 2003, and episodes are well captured throughout the year.

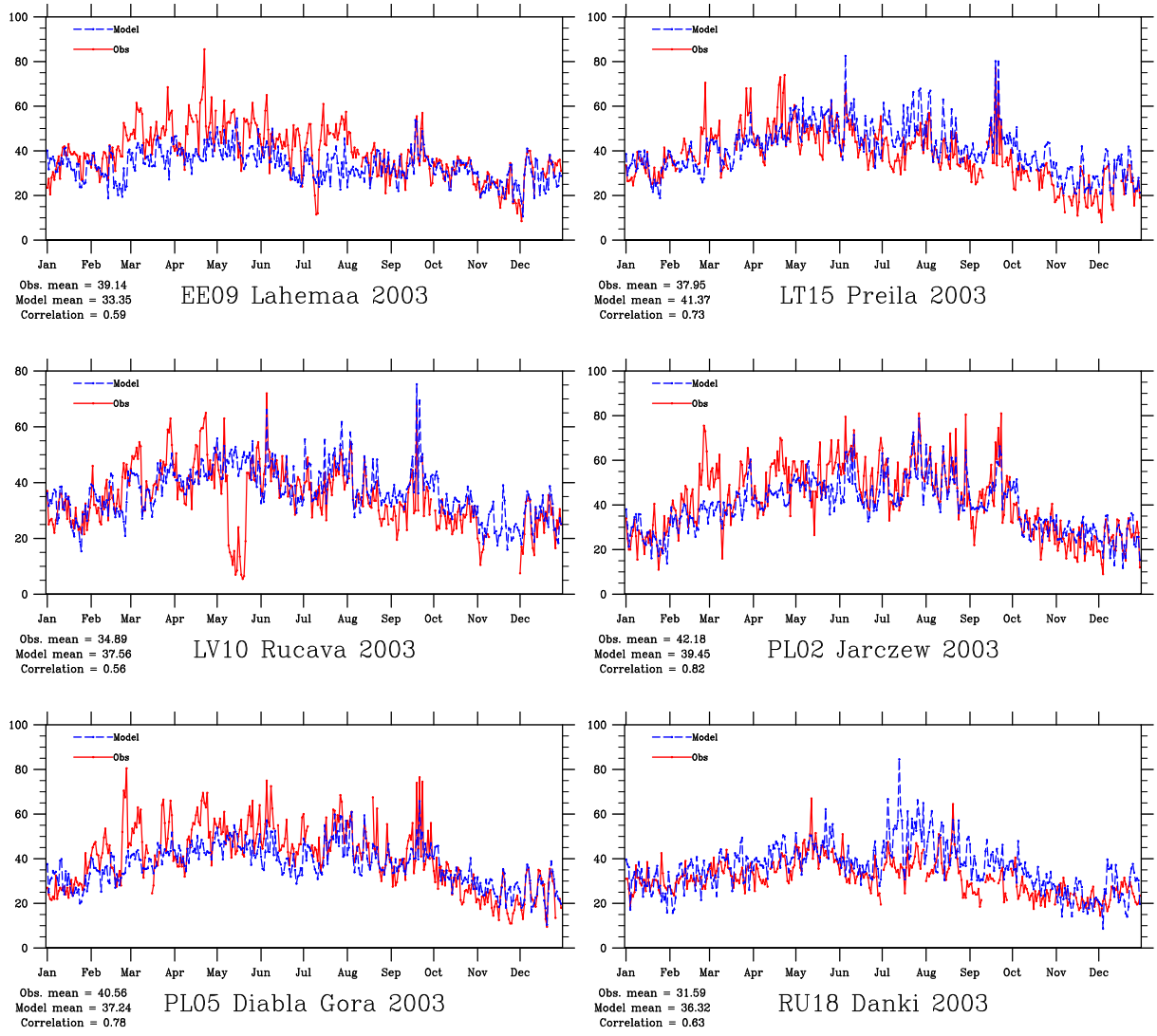

Figure 4.3: Modelled versus Observed Daily Max Ozone (ppb), North Eastern Europe, 2003

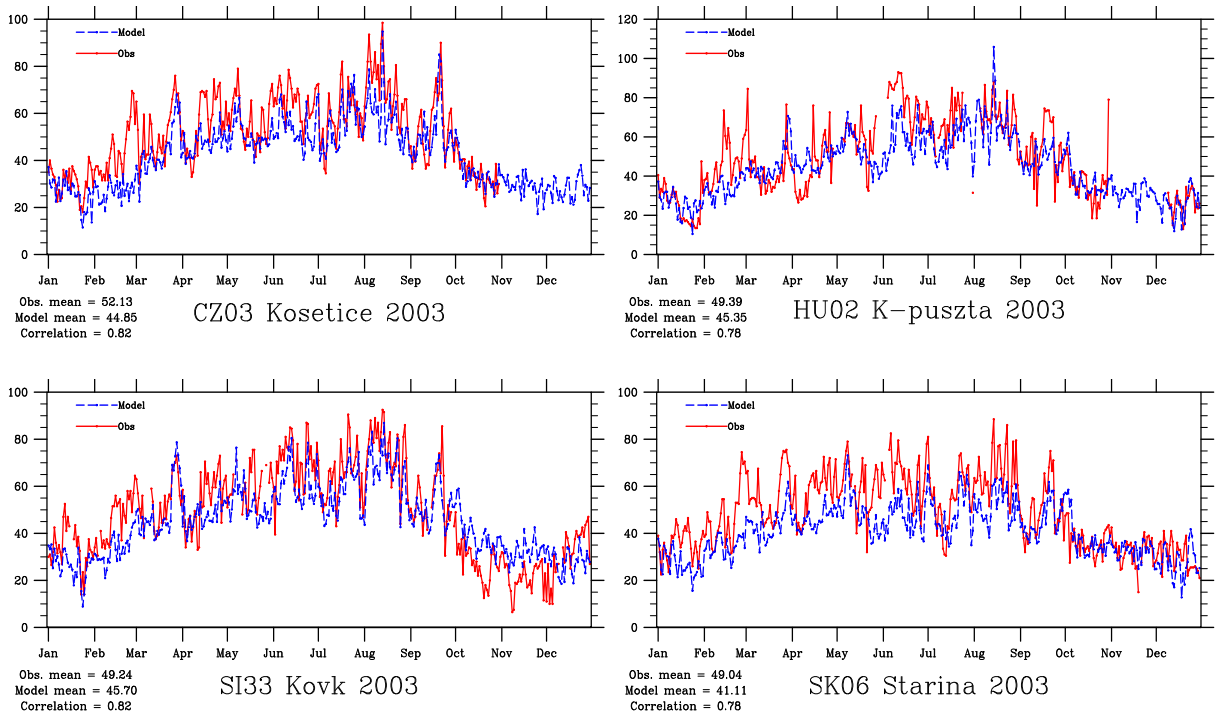

Figure 4.4: Modelled versus Observed Daily Max Ozone (ppb), East and South-East Europe, 2003

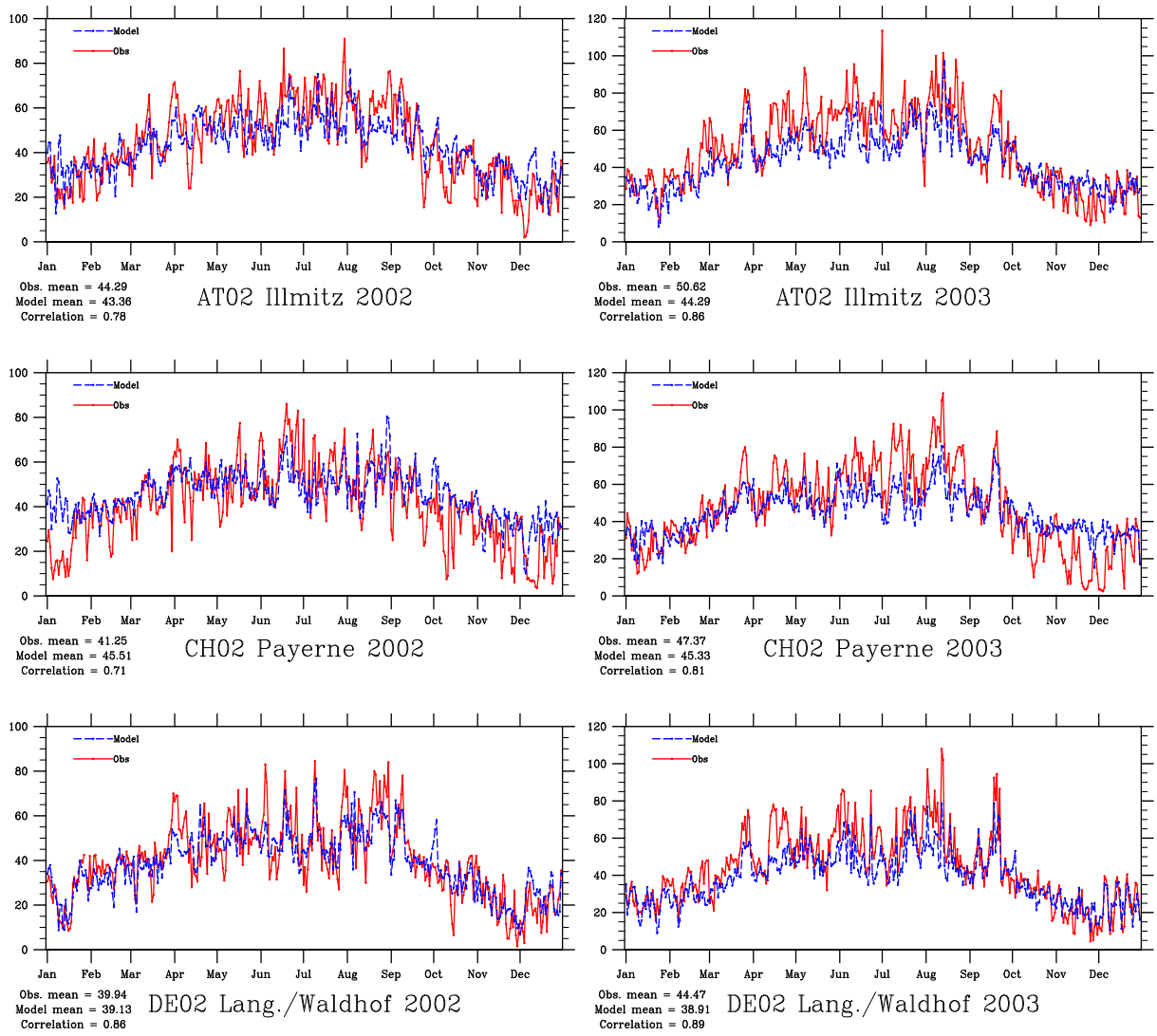

Figure 4.5: Modelled versus Observed Daily Max Ozone (ppb), Central and NW European Sites, 2002 and 2003

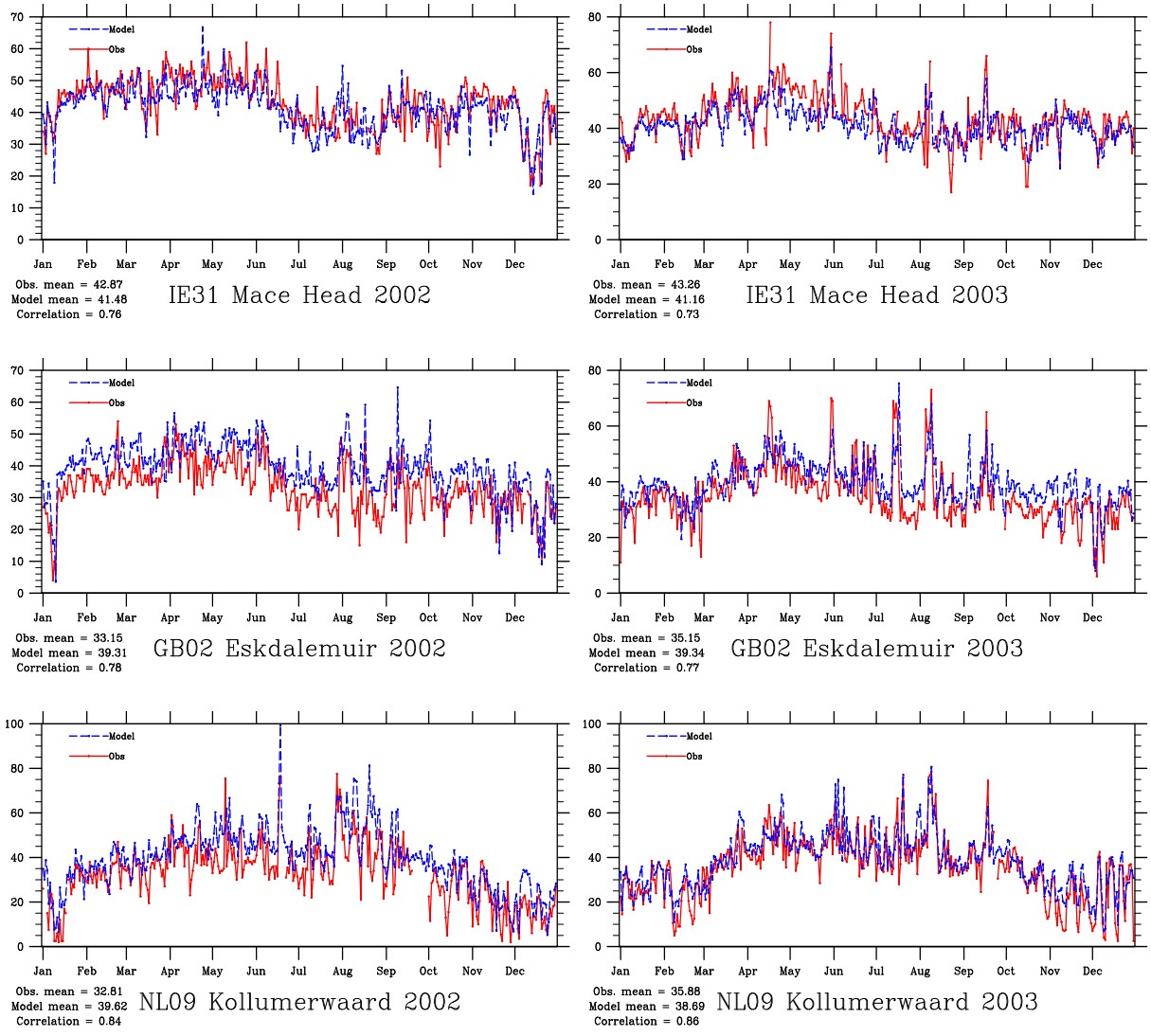

Figure 4.5: Continued

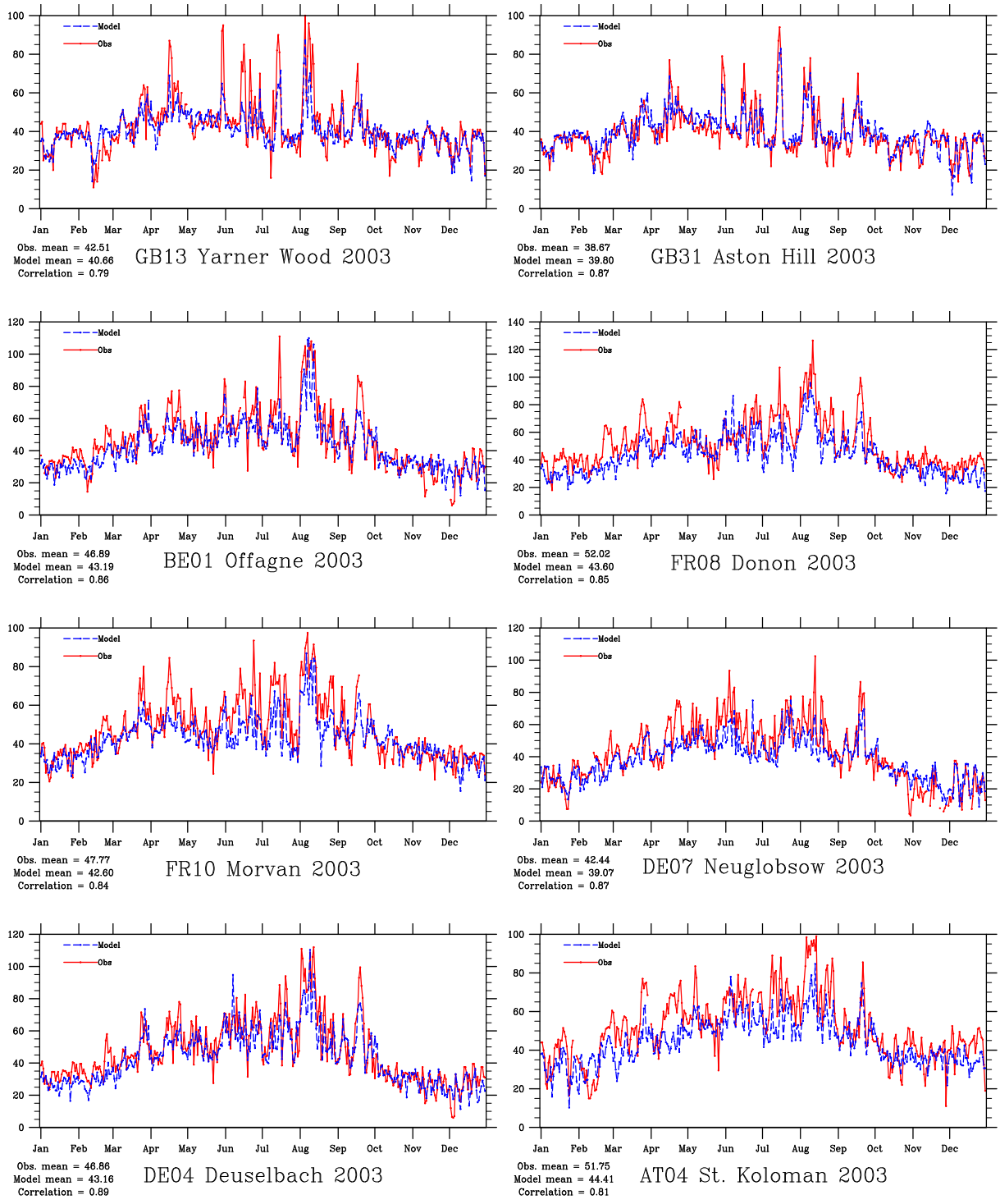

Figure 4.6: Modelled versus Observed Daily Max Ozone (ppb), Central and NW European Sites, 1999, and 2003

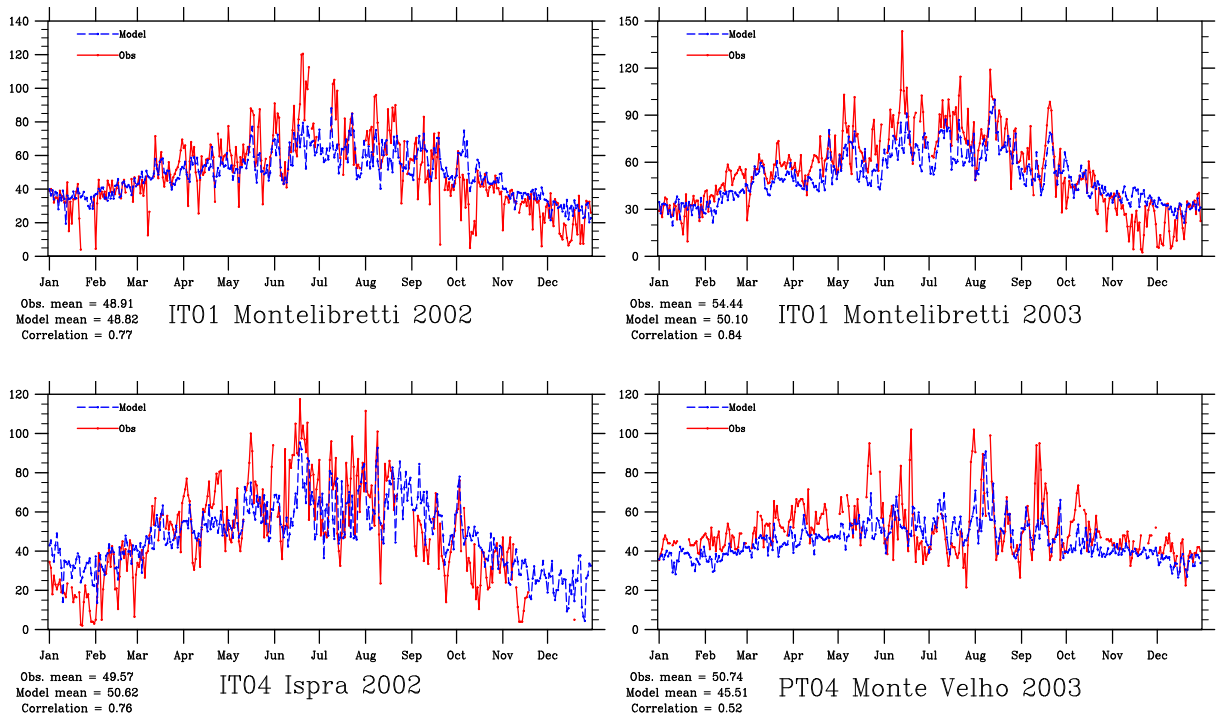

Figure 4.7: Modelled versus Observed Daily Max Ozone (ppb), Italy and Portugal

4.2.4 Mediterranean Sites and Portugal

Only one site in the Mediterranean has observations for both 2002 and 2003 – the site Montelibretti (IT01). These results are shown in Figure 4.7. Not surprisingly the plots for this site resemble those for the nearby Swiss and Austrian sites - with better correlations in 2003 but greater bias (underestimation) also. It can be noted that Montelibretti is situated near to Rome, and often influenced by the urban plume – as is evident presumably in the frequent episodes of almost-zero ozone seen in December 2003.

The remaining site plots show a rather mixed performance. The French sites Iraty (FR12, close to the Spanish border) shows data-capture problems and large bias. However, Peyrusse Vieille (FR13, also southern France) shows good results throughout the year. The Spanish sites also show quite good results, although the peaks in August are underpredicted by the model.

Similarly, the long-term Portuguese site Monte Velho also shows clear signs of local influence, with ozone concentrations often depressed to low levels. This site is therefore excluded from our analysis.

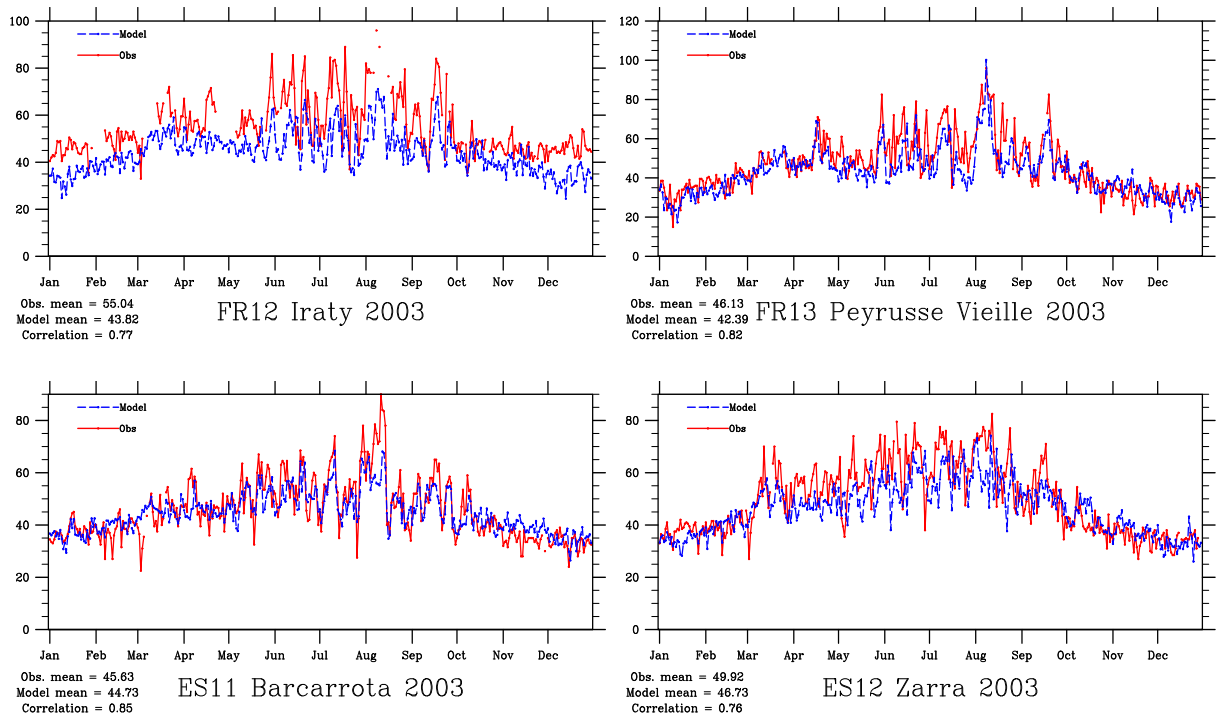

Figure 4.8: Modelled versus Observed Daily Max Ozone (ppb), Mediterranean Sites, 2003

4.3 Other statistics, O_3

The daily maximum ozone concentrations from the model and measurements can also be displayed as frequency distributions. Figure 4.9 illustrates these distributions for 2002 and 2003. Data from 127 stations are used. Three stations were excluded because of known influences from nearby sources or questionable data for 2003. The shape and peaks of the distributions are reproduced to a satisfactory degree, although the model has a clear tendency to a narrower distribution. As noted in previous reports (Simpson et al. 2003b), many reasons could be offered for such behaviour, among them the simple fact that the modelled values are averages over 50×50 km^2 grids of height $\sim$100 m, whereas the measurements are point values and hence subject to a noisier distribution. As an example, a site within a grid square is more likely to pick up local plumes of excess ozone arising from the photochemistry of nearby city emissions. On the other hand, the same site is also more likely to pick up plumes of ozone-depleted air which arise from NO_x titration, due perhaps to nearby traffic or urban area sources. It may well be that in 2003 these local plume effects are stronger, producing the poorer performance for 2003 than seen in previous comparisons.

In recent years the so-called AOT40 index has been used as an indicator of risk to vegetation. AOT40 is defined in section 1.2. As was made clear in Simpson et al.

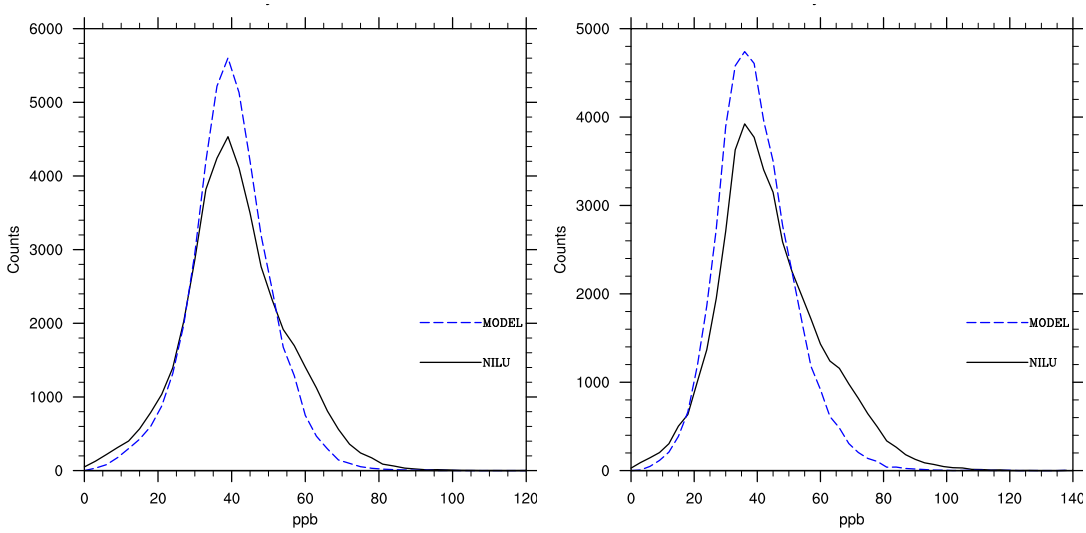

Figure 4.9: Comparison of Modelled (dashed line) versus Observed (solid line) Ozone values, 2002 (left) and 2003 (right). Frequency distribution derived from all daily maximum ozone values for 127 stations.

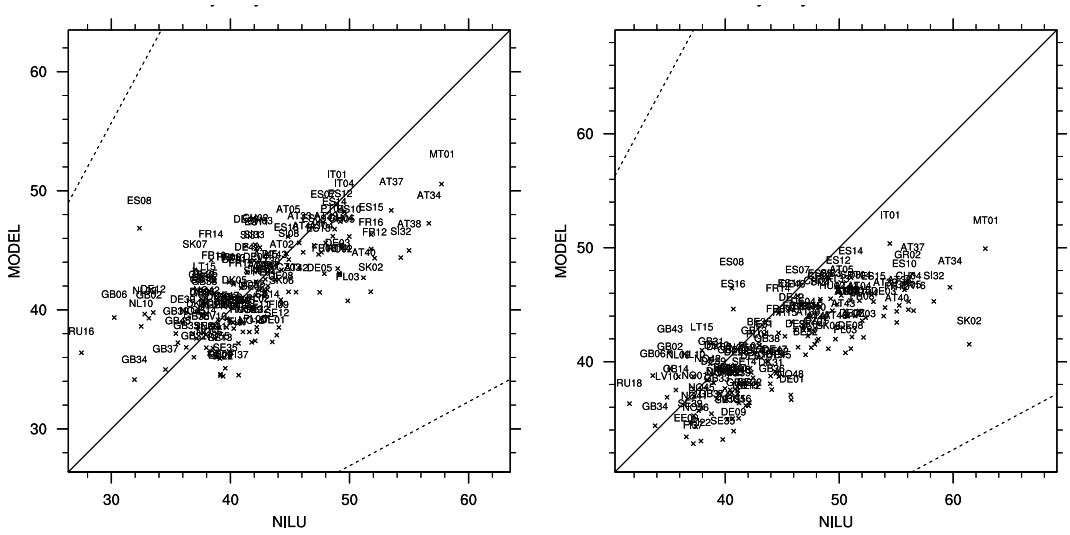

Figure 4.10: Comparison of Modelled versus Observed Daily Max. Ozone values, 2002 (left) and 2003 (right)

(1998), AOT40 and the similarly defined AOT60 have the disadvantage that they are very sensitive to small systematic errors in either the model or the measurements. Such errors are unavoidable and have many sources. Observations of ozone should usually have a good accuracy and precision with modern instruments, but calibration procedures are not uniform across the EMEP network and uncertainties of at least $\pm 5\%$ seem very likely. In addition, measurements are affected by site placement and local

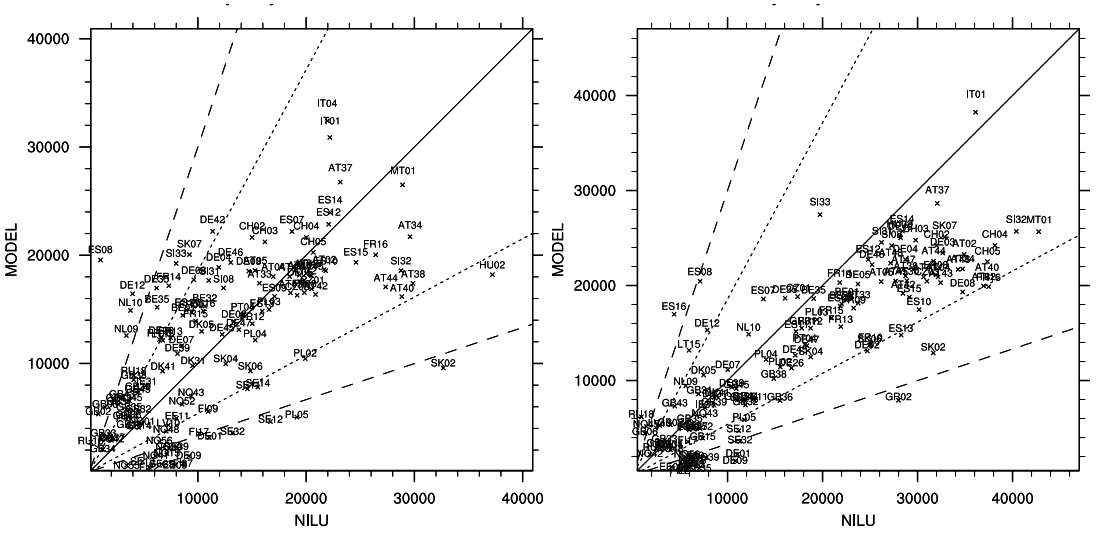

Figure 4.11: Comparison of Modelled versus Observed AOT40f values, 2002 (left) and 2003 (right)

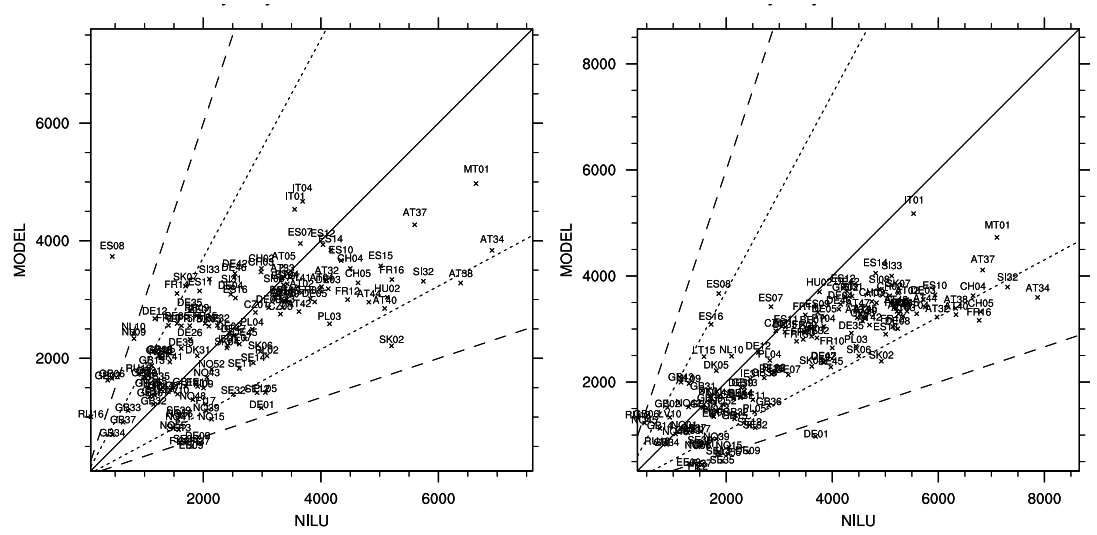

Figure 4.12: Comparison of Modelled versus Observed SOMO35 values, 2002 (left) and 2003 (right)

NO_x sources, so reported ozone values cannot represent the perfect regional average for an EMEP grid square. Sensitivity studies performed in both Simpson et al. (1998) and Tuovinen (2000) showed that uncertainties in AOT40 can be very significant, especially for sites where ozone levels often lie around the 40 ppb threshold value.

However, AOT40 is still an important statistic, and we present here comparisons between the modelled and observed values.

Figure 4.11 shows the observed and modelled $AOT40_f$ values for the years 2002

and 2003 for all stations having a summer-time data-capture of $>$ 90%. (Defined here as days with more than 18 hours of hourly observations). Despite the difficulties expected for modelling this parameter, reflected in a large scatter, it seems that the model does a reasonable job of predicting AOT40 levels at most stations. In 2003 there is a clear tendency towards model underprediction, however, consistent with the under-predictions shown in the time-series analysis presented above.

For health issues, the index SOMO35 is now recommended (section 1.2). Figure 4.12 shows the observed and modelled SOMO35 values for the years 2002 and 2003 for all stations having a summer-time data-capture of $>$ 90%. (Defined here as days with more than 18 hours of hourly observations). Despite some scatter, that the model does a reasonable job of predicting SOMO35 levels at most stations, even for the extreme year 2003 (although again with a tendency for underprediction).

4.4 Nitrogen Dioxide

Nitrogen oxides (NO_x = NO + NO_2) are arguably the most important precursor for photochemical oxidant formation. In rural atmospheres NO_x consists largely of nitrogen dioxide, and measurements of this compound have been conducted in the EMEP networks since 1980. These measurements consist of daily mean values, sampled from 6 GMT of one day to 6 GMT of the next, so modelled concentrations have been averaged over the same time-period.

It should be noted that NO_2 is a much more difficult pollutant to predict than ozone. Partly this is because NO_2 is a gas whose concentrations are often dominated by ground-level sources, especially traffic. Nearby roads or urban areas can therefore influence measurements in a way which a model with 50×50 km^2 resolution, and with a lowest layer of approximately 90 m depth, can never achieve. Concentrations of NO_2 are also very sensitive to uncertainties in some chemical parameters (notably the concentration of the OH radical) and to meteorology (stability, dispersion). Finally, measurements of NO_2 may be subject to interferences from other gases, especially at the very low ($\leq$ 1 ppb) concentrations often found at EMEP sites. This is not usually a problem when using the reference method, NaI impregnated glass sinters, but may affect some sites which used other methods for some years.

Figure 4.13 presents scatter-plots of modelled versus observed NO_2 for the years 2002 and 2003. These plots show the 1:1 line (solid), the line of $\pm$30% bias, and the lines of $\pm$50% bias. For both 2002 and 2003 most sites ($>$70%) fall within the $\pm$30% lines, and around 90% fall within the 50% bias line. There are some outliers in both years, partly associated with site location. For a primary pollutant such as NO_2 this is probably not surprising, and can possibly be explained by nearby NO_x sources influencing the measurements. However, even including these outliers in the analysis, the overall mean bias of -23% (for 2000) is very satisfactory. The correlation coefficient for 2003 (r =0.54) is significantly worse than that for 2002 (r =0.75), and the reasons for this remain to be explored.

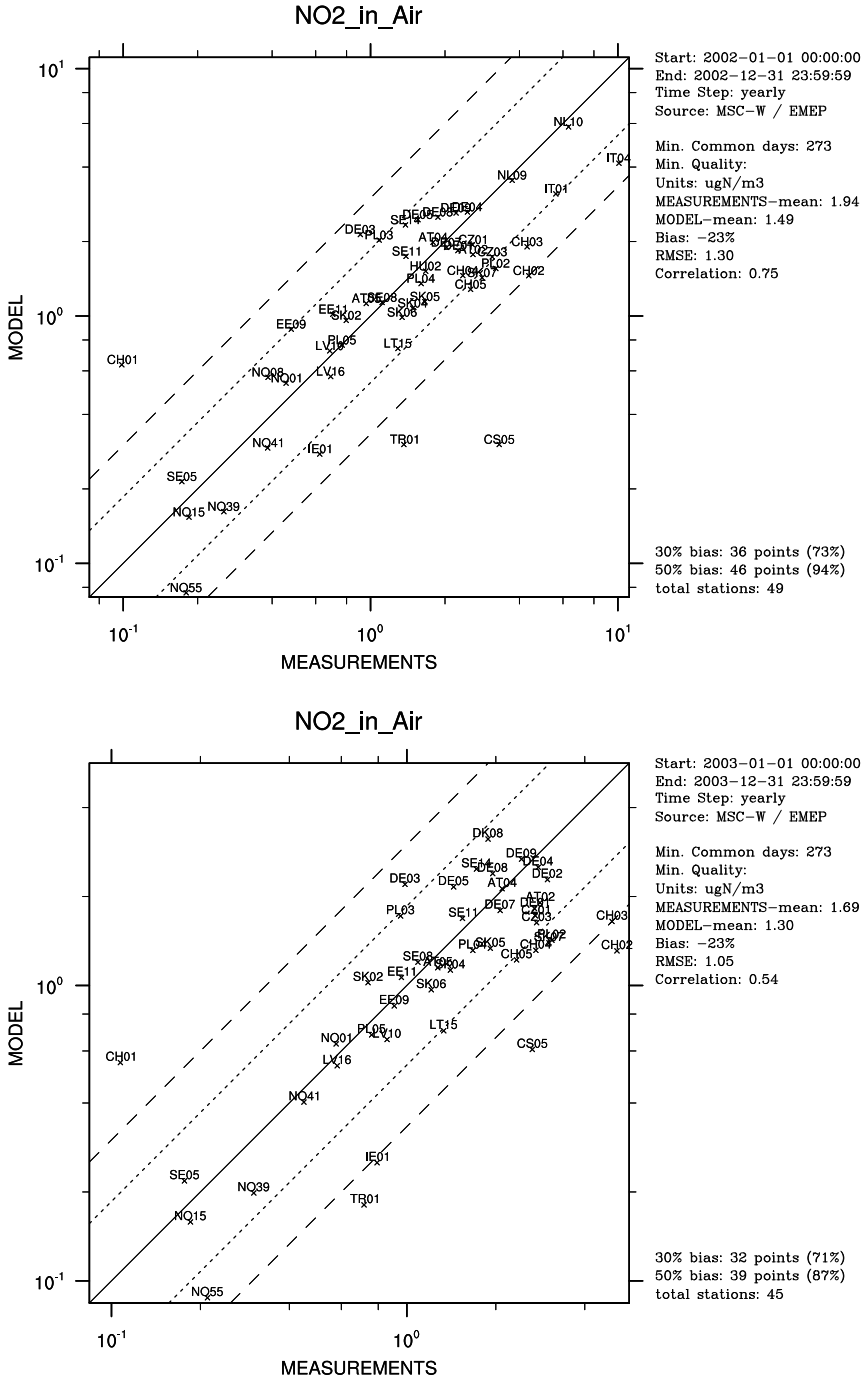

Figure 4.13: Comparison of Modelled versus Observed Annual NO_2 Concentrations, Year 2002 (top) and 2003 (bottom).

4.5 Discussion and Conclusions

This chapter has presented the performance of the EMEP MSC-W Eulerian model for ozone, and to some extent, NO_2 for 2002 and 2003. The general findings are that the model performs better for 2002 than for the year 2003 in terms of mean ozone, AOT40 and SOMO35, but that correlation coefficients were higher in 2003, often with $r > 0.8$.

There are many possible reasons for the under-predictions found in the summer of 2003, and also in in the springtime of 2003. Possible reasons may include problems in specifying boundary conditions (BCs). For ozone the use of Mace-Head in the correcting BCs (Simpson et al. 2003a) should avoid major errors, but no such procedure is used for other ozone precursors, for example CO. Further, the sumer of 2003 experienced very low stratospheric ozone (Orsolini and Nikulin 2005), which is not accounted for in the model. One important factor is also likely to be the lack of soil moisture effects in the standard EMEP model. As shown in Emberson et al. (2000), Simpson et al. (2003c) soil moisture deficits can induce large reductions in the deposition velocity of ozone, and this should lead to higher ozone concentrations. Unfortunately, soil water is difficult to predict reliably on the European scale, and soil water pressure which drives stomatal conductance is extremely difficult to parameterise, even on local scales. Work is ongoing in cooperation with the Stockholm Environment Institute at York (SEI-Y) to try to make progress in this important areas.

A surprising feature of 2003 is also the occurrence of high ozone levels in the springtime

It is clear that further investigation of the 2003 results is warranted, and this work will be reported in a future publication.

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