Hilatar, a limited area simulation model of acid contaminants

Part II Long-term simulation results

Atmospheric Environment 37/11 pp. 1535-1547

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Abstract

This paper summarises multi-annual simulation studies with the Eulerian transport-chemistry model Hilatar over Europe and the Baltic Sea region. The modelled concentration and deposition fields and model-measurement intercomparison results are presented and variability patterns are analysed. Although the European emissions slightly decreased during the study period, no clear decreasing trend in deposition was detected over any sub-area surrounding the Baltic Sea, partly due to the strong inter-annual meteorological variability.

The study has generated a continuously updated data base for environmental research projects containing gridded fluxes and concentrations over Europe.

Keywords: European modelling, deposition, nitrogen, sulphur, model-measurement comparison

INTRODUCTION

3D models have been successfully used for estimating long-term pollution over Europe (see Zlatev et al., 2001 for Nordic countries). However, impact studies would greatly benefit from data bases of resulting fields with high time and space resolution over several years, since elevated air concentrations and deposition events occur episodically and often simultaneously with extreme meteorological conditions. Monitoring at Finnish background stations shows that up to 90 % of the monthly wet deposition can be received during one single day that is not necessarily the most rainy of the month (Leinonen 1994-2001, Ruoho-Airola & Salmi, 2001). The response of a living organism is not the same if annual or monthly averages instead of instant exposure peaks or deposition loads together with meteorological stress are used in damage studies.

We present here long-term simulation results of the model described in the first part of the article Hongisto (2002), together with model-measurement comparisons over the whole European domain. The results were generated over the period 1993-1998 for the EU-BASYS (Baltic Sea System Study) subproject 5: Air pollution load to the Baltic Sea. More model results are presented on http://www.fmi.fi/research_air/air_25.html.

MODEL STRUCTURE AND INPUTS

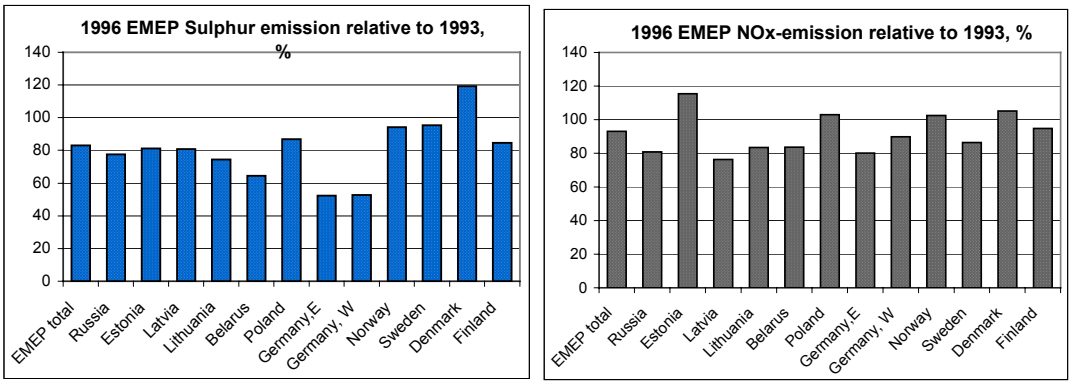
The Hilatar model (Hongisto, 1998), described in Part I of this article, covers either the whole of Europe (referred here as the EUR-Hilatar), the Baltic Sea drainage basin with its

surroundings (referred as the NMR-model) or Finland, with horizontal grid resolution varying from 56 to 11 km. The meteorological input is taken from the 6-hour predictions of the operational weather forecast model HIRLAM of the Finnish Meteorological Institute (FMI).

We use the EMEP 50-km gridded emission inventory (www.emep.int), merged in the NMR-area with the Finnish, Russian and Estonian stack inventories and areal emissions (Häkkinen et al., 1995; Pietarila et al., 1996; Mäkelä & Salo, 1994) updated with additional statistical information. Effective plume height is estimated for the stacks, emission-specific vertical height profiles are used for areal sources. Country-specific daily emission indices from the GENEMIS project (Lenhart et al., 1997) are used together with monthly, daily and diurnal indices of the FMI local emission inventory.

Over the NMR-domain, about 20 % of the 1993 sulphur emissions were located in the Kola Peninsula, 26 % in Estonia and Russia, 5 % in Finland and over 48 % in the southern part of the model area. Only about 2 % of oxidised nitrogen is emitted in the Kola Peninsula (with 1.12 M inhabitants), 10 % in Estonia and Russia, in comparison with 14 % in Finland.

The European SO_2-emissions decreased from 1993 to 1996 by 15 %, NO_2 by 7 % and NH_3 by 5 %, but this decrease was not even over all countries (Fig. 1). Annual differences are temperature dependent: emissions increased in the Nordic countries during the cold year 1996 compared to 1995. NH_3-emissions decreased most in the East-European countries, even by 45% in Lithuania.



Fig, 1: Emission changes (%) between 1993 and 1996 in countries around the Baltic Sea.

RESULTS OF 6-YEARS SIMULATIONS

Areal deposition distributions

Over the NMR-domain the 1993-1998 average total nitrogen and sulphur deposition distributions (Figs. 2 - 3) have the expected North-South gradient. It is superimposed by an east-west gradient for sulphur due to north-western Russian sources. The European sulphur deposition exceeded 2.5 g m^{-2} in the most loaded, the Black Triangle area.

The total deposition of nitrogen over the Baltic Sea (Fig. 2) is about 1.5 times higher in winter and autumn than in summer and spring due to seasonal variation in precipitation and emissions. Some nitrogen compounds are efficiently scavenged by snow. Over water, total deposition consists mainly of wet deposition, however in South Sweden and on Danish Islands in summer the dry deposition share of the total nitrogen deposition exceeded 50% due to high summertime ammonium emissions. The dry deposition share decreased northwards and in

high altitude areas, being the smallest (<10%) in winter over the Kola Peninsula and over parts of the Scandinavian mountains.

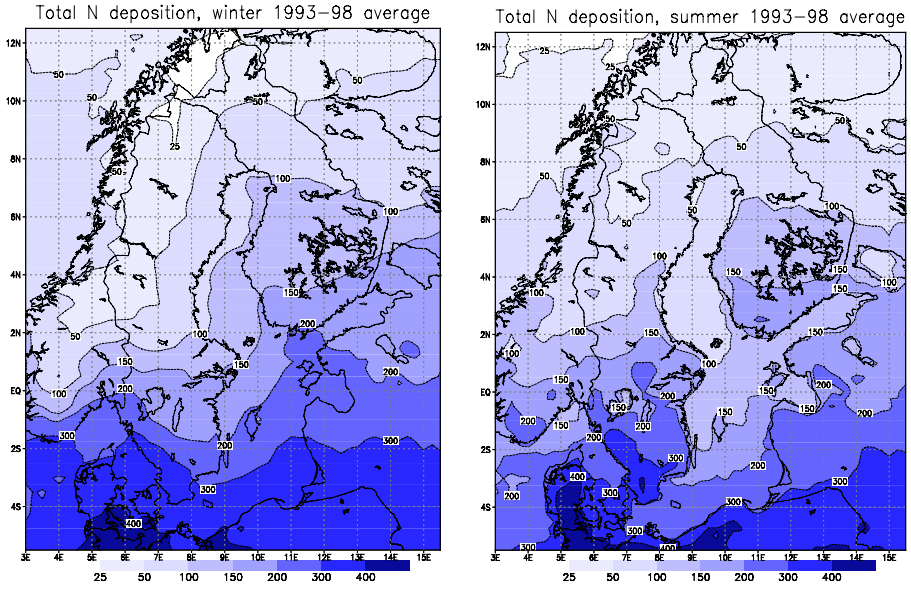


Fig. 2: Total nitrogen, 1993-1998 winter and summer 3 month deposition averages (mg(N) m^{-2}).

The relative standard deviation of the monthly S deposition is presented in Fig. 4. Similarly, the std of the N deposition was highest over the Atlantic in winter or spring months (it could exceed 100%) or in remote or mountainous areas. This is due to geographically uneven source distribution of nitrogen, since a remote locations can receive either clean or polluted air over long period depending on the prevailing wind direction. Additionally, the variability of mixing conditions over water is strongly dependent on the strength of the winter and the ice coverage.

Over the NMR-area, the highest absolute variation was found over coastal and southern areas. The inter-annual variation of the monthly wet deposition was the highest over the North Atlantic and the Norwegian coast and during summertime also over the Baltic Sea.

Seasonal variation of sulphur deposition was higher than that of nitrogen in accordance with the variation of the sulphur dry deposition share and the more pronounced seasonal variation of emission intensity. Over the NMR-domain the S dry share was the highest over the Southern Baltic Sea (exceeding frequently 60% except in spring) and below 10% most of the time over the other northern areas except the Kola Peninsula. The variability of S-deposition is very large close to some strong stack sources.

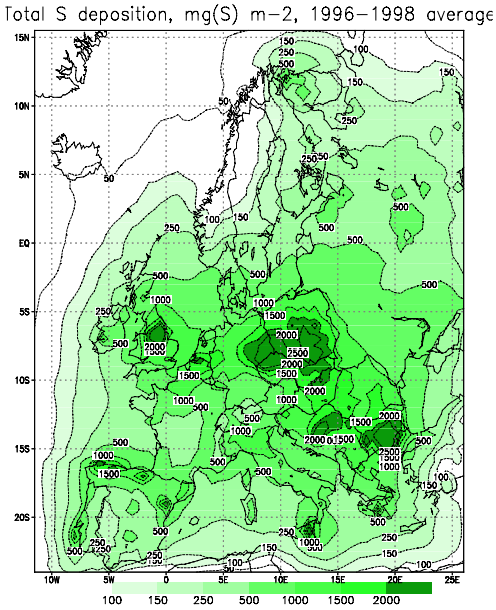


Fig. 3: Total average sulphur deposition (mg(S) m^{-2}) and its 1993-1998 monthly standard deviation over the EUR-model domain.

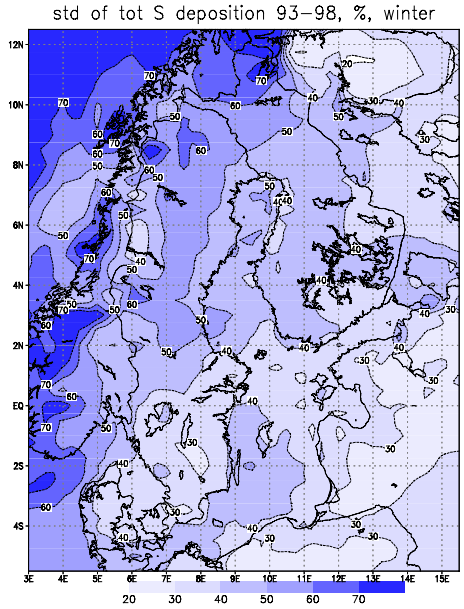


Fig 4. 1993-1998 monthly standard deviation of the sulphur deposition, %, over the NMR-model domain.

Inter-annual variation of deposition

The regional interannual variability of the NOx, NHx and S deposition during the period 1993-1998 was high (Fig.5). The selected regions were seven land areas and five Baltic Sea sub-areas. Sulphur deposition was generally the highest during cold winters and spring 94 and 96. The slight reduction of nitrogen emission is not reflected in the deposition values for any area. Prevailing meteorological conditions affected more the annual regional variation. For instance, NOx-deposition was minimal in 1995 over Denmark due to low precipitation during spring and summer, and in 1996 over the Baltic States due to low winter precipitation.

Trends for sulphur and total nitrogen (oxidised+reduced) over the whole of Europe as modelled by the EMEP model (EMEP, 2000) display a slight decreasing trend between 1993 and 1996 and an ascending one afterwards until 1998, a feature that can be seen in our Fig. 5.

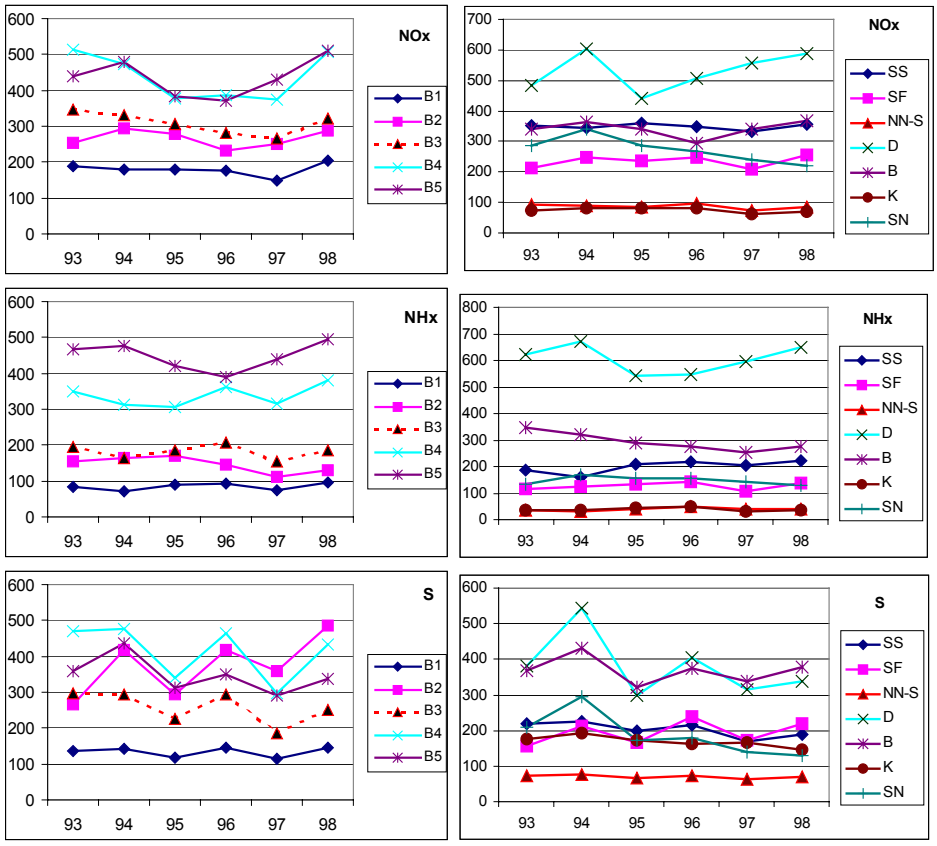


Fig 5. Inter-annual variation of modelled regional deposition of oxidised nitrogen, reduced nitrogen and sulphur, (tons grid^{-1} yr^{-1}) for different Baltic Sea Basins (left panel) and land areas (right panel). The area codes are SS: Southern Sweden, SF: Southern Finland, NN-S: Northern Norway and Sweden, D: Denmark, B: Baltia, K: Kola Peninsula, SN: Southern Norway, and: B1: Gulf of Bothnia, B2: Gulf of Finland, B3: North Baltic Proper, B4: South Baltic Proper and B5: Kattegat/Belt Sea.

Concentrations

Concentrations of nitrogen compounds at the lowest model level decrease northwards with increasing travel distance from the most intensive source areas (Fig. 6). We present the winter SO_2-concentration fields because critical values are defined for this period. Modelled sulphur concentrations have high peaks also in the vicinity of northern and eastern sources. Oxidised nitrogen and sulphur concentrations are the highest during winter due to maximum emissions and frequent inversions. The summertime daily average atmospheric boundary layer (ABL) height is more than six-times higher than in winter over land, although a minimum ABL height of 150 m was set. Ammonia emissions are the highest during summer and its low concentration is more effectively converted to ammonium-sulphate particles in winter when there is more sulphate in air. HNO_3 levels are low in winter due to low OH concentrations, short days and temperature-independent reaction with NH_3 producing ammonium nitrate near the surface.

Over the NMR-model domain, NO_x and SO_2 winter concentrations decrease until 1995, increasing in 1996. During summer, NO_x concentration exceeds the period average in 1997-1998 in the eastern parts of the model, but in the western parts in 1993, 1994 and 1996. Particulate concentrations at NMR-model boundaries were the highest in 1993-1994 reflecting the fact that secondary substances calculated with the EMEP one-layer model are closer to the

surface than in the EUR-Hilatar. However, particle concentrations were higher during the cold year 1996 (with EUR-Hilatar boundaries) than in 1995 (with EMEP boundaries). Generally no trend can be detected, the concentrations of individual compounds over a specific geographical area seem to depend on meteorology: precipitation, main advection direction relative to the main source areas, cloudiness and availability of reactants.

In Europe (Council directive 1999/30/EC), limit values for vegetation in background areas are: 20 μg m^{-3} for SO_2 during winter, 30 μg m^{-3} for NO_x annual average concentration. Critical values for SO_2 are exceeded in winter 1996 over large areas in Central Europe (broad Black Triangle area, Hungary) but also over the Balkan countries, as well as the Midlands in UK and the western Po Valley. The same overall picture occurred in 1997 and 1998 but the exceedence areas were smaller and more scattered. Limit values for NO_2 were not exceeded during those years.

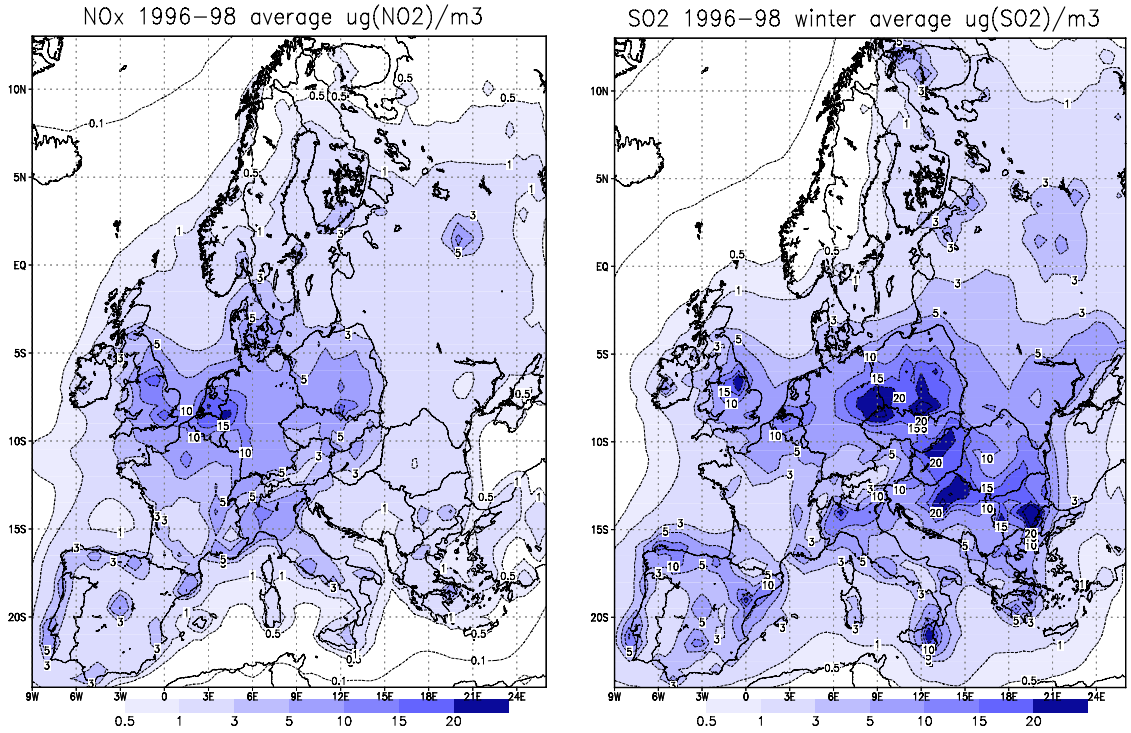


Fig. 6. Modelled 1996-1998 average annual NO_2 and winter SO_2 concentrations in Europe.

Vertical profiles of concentrations

The influence of the simultaneous change of the NMR-model boundary values from the 1-layer EMEP to 3-D European Hilatar values, and of the rise of the top of the model from 3 to 10 km were studied by comparing inter-annual variation of mass and concentration profiles averaged over selected areas. The detected inter-annual changes in the profiles were sometimes opposite over different geographical area, depending mainly on inter-annual variation in meteorology and were almost negligible for primary pollutants. The shape of the profiles is mainly determined by meteorology and emissions. Summertime convection over land and wintertime mixing over the open water are the strongest processes. Leakage across the mixing height as well as vertical advection are weak.

Above 3 km the mass share of SO_2 was 0.1% of the total vertically integrated mass in winter and below 0.3% in summer. Over Southern Scandinavia and Finland, around 90% of the vertical NO_x and NH_3 mass, 80% of the SO_2 mass and 50-70% of the HNO_3 mass is below 900 m, while over Northern areas this share is 10% lower for all compounds. It is expected

that raising the upper boundary of the model will not dramatically affect the results. The fraction of secondary species increases in aged air masses and with altitude. The vertical total sulphur mass above most of the sub-areas was the highest in 1996, when the sink processes were the slowest.

Relative concentration profiles, weighted by the mean along the vertical, were generally sharper near the ground over sea than over land. In summer, ground-emitted compounds reached high layers over land. Nitric acid has the strongest chemical and dry deposition sinks near the surface, and its annual relative concentration is 1.5 …6-times higher at levels 4-5 (600-900 m) than near the surface. The strongest absolute concentration increase with height can be seen in areas with frequent inversions located downwind of stack emission sources, or when the surface sink is strong, e.g., over open sea areas in winter.

The one-layer EMEP-model concentrations were split into different layers by height profiles at the NMR-model boundaries, and a weak mixing across the ABL height was allowed. In the EUR-Hilatar, emission height profiles are used and the mixing occurs either into the layers 1-3 near the surface (below 200 m, low level sources), or, in case of high level sources, mainly into the layers 3-4 above the surface, main part below 650 m. The pollutants are lifted up by vertical advection or turbulence yielding to realistic profiles. On the other hand, in the EMEP-model, the mixing height is generally higher than that in the EUR-Hilatar, leading to more diluted concentrations.

MODEL-MEASUREMENT COMPARISON

The accuracy and consistency of the results were assessed by comparing the modelled and measured values, using the statistical package of Sofiev (1999) in the EUR model domain (0.5^o grid averages against about 90 EMEP-station data). The Hilatar post-processing routines (Hongisto, 1998) were used for comparison of the NMR-0.25^o model results at 29 EMEP stations. Daily concentrations of SO_2, NO_2, NH_3, $SO_4^=$, NO_3^-, NH_4^+, HNO_3+NO_3^- and NH_3+NH_4^+ in air, and monthly mean wet deposition of $SO_4^=$, NO_3^- and NH_4^+ were compared with the EMEP measurements (www.emep.int) over the period 1993-1998. All sites located inside the model domain were included in the comparison with no a-priori filtration of the data.

For almost all species the EUR-model demonstrated quite good agreement with measurements over Central and Northern Europe, while for Southern Europe the results are usually not so encouraging (Fig. 7). The "good-correspondence area" varies from one species to another and covers in the best case: Germany, Western Poland, France, the Benelux, UK, the Baltic region and most of Scandinavia. One reason is, that the density and accuracy of observations and emission inventories in Southern Europe is lower (e.g., ship emissions for the Mediterranean area were missing at the time of the simulations). The best performance of the NMR-model was in South-Western areas of the domain, i.e., southern Scandinavia and Finland.

Oxidised nitrogen

In the EUR-model, NO_x concentrations are somewhat underestimated (Fig 7) over Scandinavia, Germany and UK. At some stations the correspondence is almost 1:1. Within the NMR-area, underestimation is most pronounced at coastal stations and close to emission areas during inversion situations, because the minimum mixing height was set to 150 m and the low level emissions were initially mixed through at least the two lowest model layers. At some stations e.g. when the measured concentrations are very low, NO_x was overestimated. The reasons are e.g. a too low HIRLAM mixing height or the splitting up method discussed in the

previous part I. Daily correlation exceeded 0.5 during more than 40% of the months at 9 stations, while it was worse at eastern and remote stations.

The correlation of the sum $HNO_3+NO_3^-$ is above 0.5 over the Baltic Sea region and UK. Measurements in other parts of Europe were too scarce for any firm conclusions. The absolute level of concentrations is overestimated by 30-100%. In the NMR-domain the overestimation increases northwards although average concentrations decrease to 10% of the level at southern stations. The seasonal variation of nitrate is stronger than that measured.

Nitrogen wet deposition was overestimated by 30 % over most of the European domain mainly during small deposition events, and underestimated at a few stations spread out over Europe. The difference was higher in Southern Europe. The temporal correlation was considerably better than that for air concentrations. It exceeds 0.5 for more than half of the stations. Systematic low correlation appears only in Southern Europe.

Over the NMR-area, NOx wet deposition was well predicted at Danish and German stations but overestimated in Finland and LT-15 (Preila) in the southern Baltic, and at RU-1 at the northern Finnish-Russian border. But, NO_x deposition measured at Finnish EMEP stations by FMI is systematically lower than at stations of the Finnish Environmental Institute (Vuorenmaa et al., 2000). Along the North Sea coast and at Hoburg, Gotland, maximum deposition months are underestimated, maybe due to incomplete ship-emission inventory, as well as missing non-European contribution and natural emissions.

Sulphur compounds.

With the EUR-model, SO_2 is somehow overestimated over Central Europe (by ~0.5-2 $\mu g(S)m^{-3}$) and underestimated in Northern Scandinavia and Northern England. The correlation coefficient for daily SO_2 reaches 0.4–0.6 in summer and over 0.6 in winter over the "good correspondence area". These values are quite stable (the standard deviation of the correlation coefficient is less than 20%) and close to the maximum possible level of ~0.7-0.8 for daily averaging, which is determined by the representativeness error discussed below. The concentration of sulphate in aerosol is generally underestimated everywhere by ~0.2-0.5 $\mu g(S)m^{-3}$, except over the Black Triangle region, where an overestimation of 0.3 $\mu g(S)m^{-3}$ appears. Daily correlation is nearly the same as for SO_2 and generally exceeds 0.6 over Scandinavia, UK, most of Germany and Benelux.

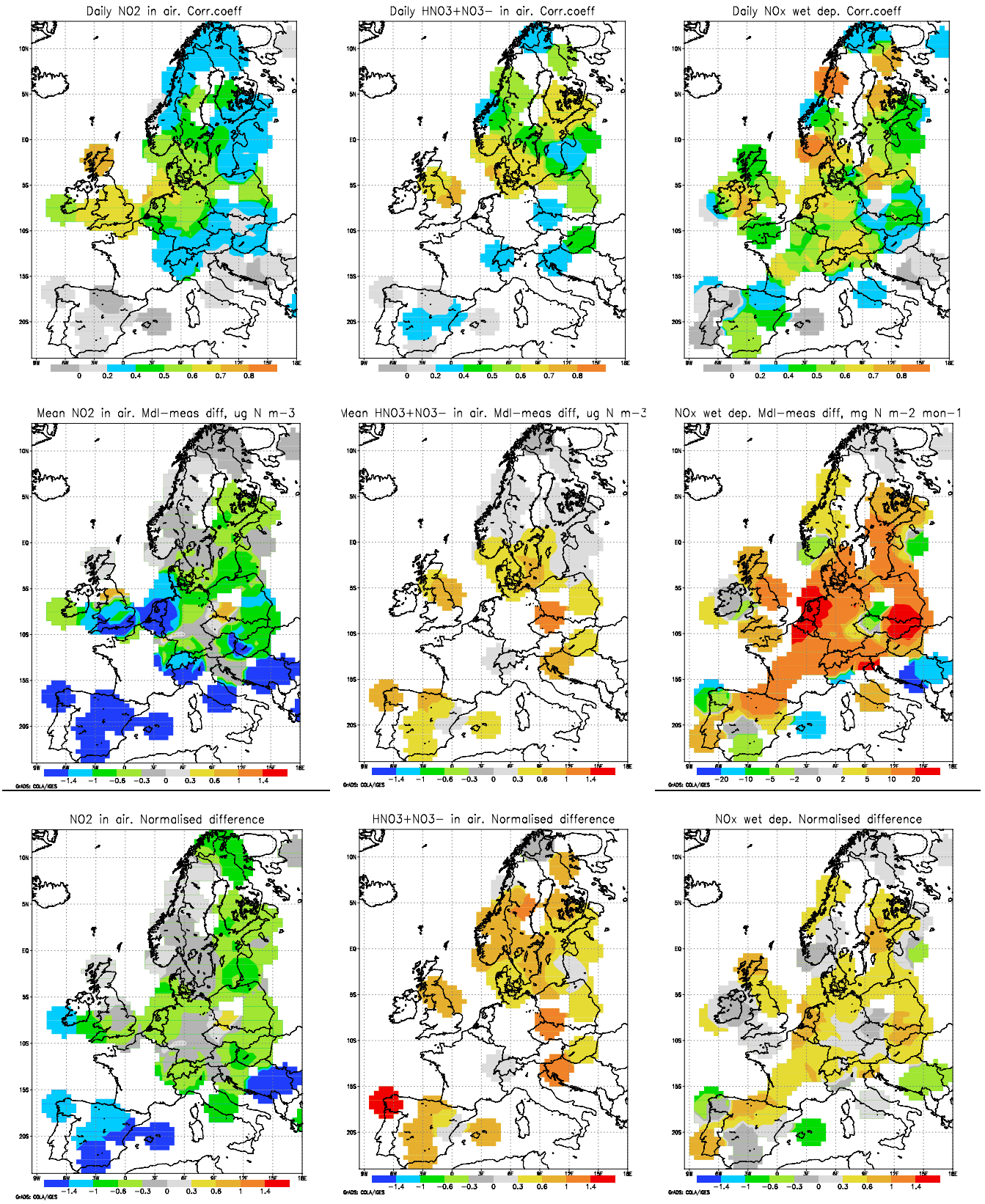


Fig. 7a: Upper panel: Spatial distribution of correlation between modelled and measured daily NO_2 and HNO_3+NO_3 and monthly NOx wet deposition. Middle and lower panels: Spatial distribution of the absolute differences and the weighted absolute difference (defined as {c(model) - c(meas)}/0.5*{ c(model) - c(meas)}, unitless) of the same compounds.

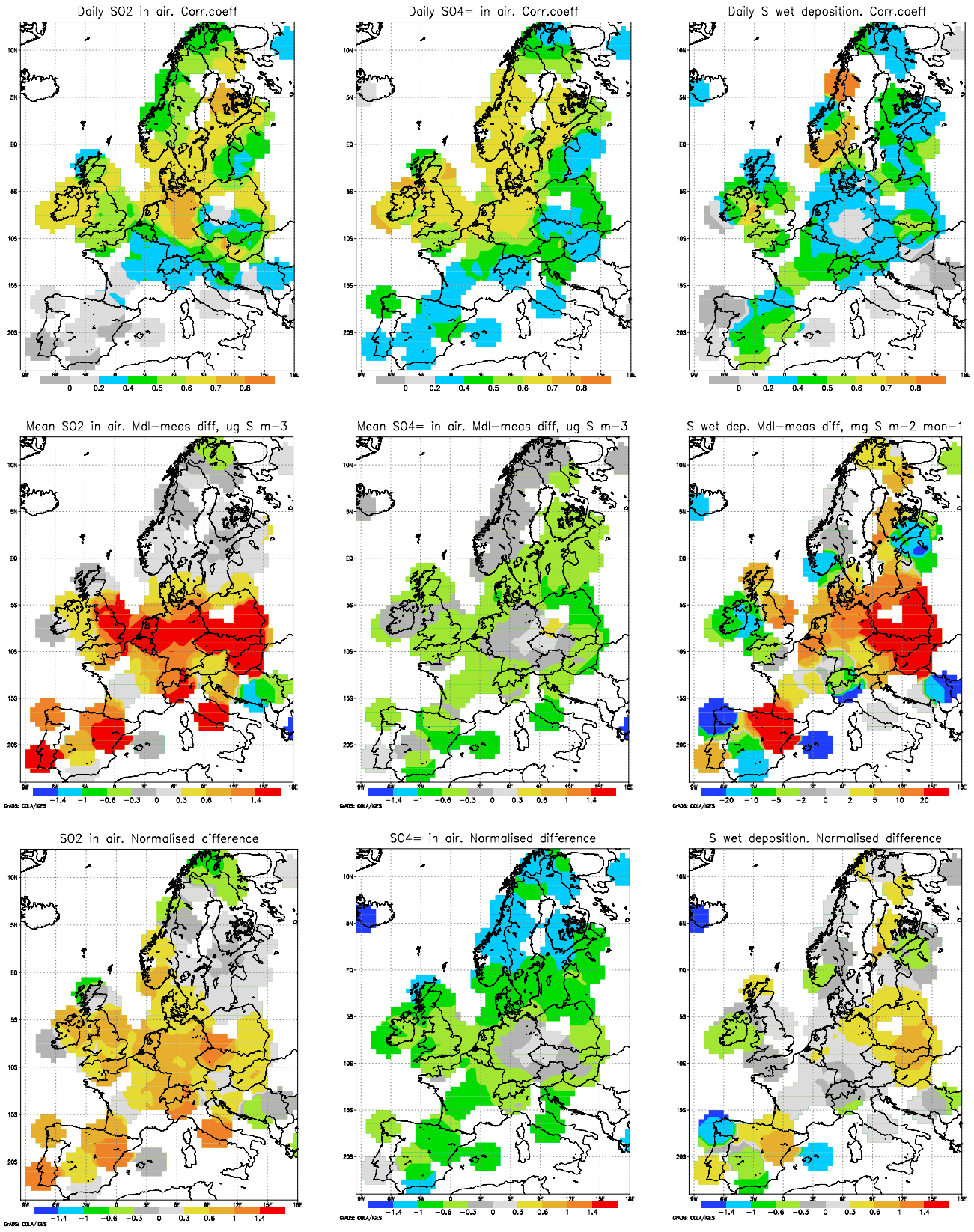


Fig. 7b: Upper panel: Spatial distribution of correlation between modelled and measured daily SO_2, SO_4 in air and monthly SOx wet deposition. Middle and lower panels: Spatial Distribution of the absolute differences and the weighted absolute difference (unitless) of the same compounds.

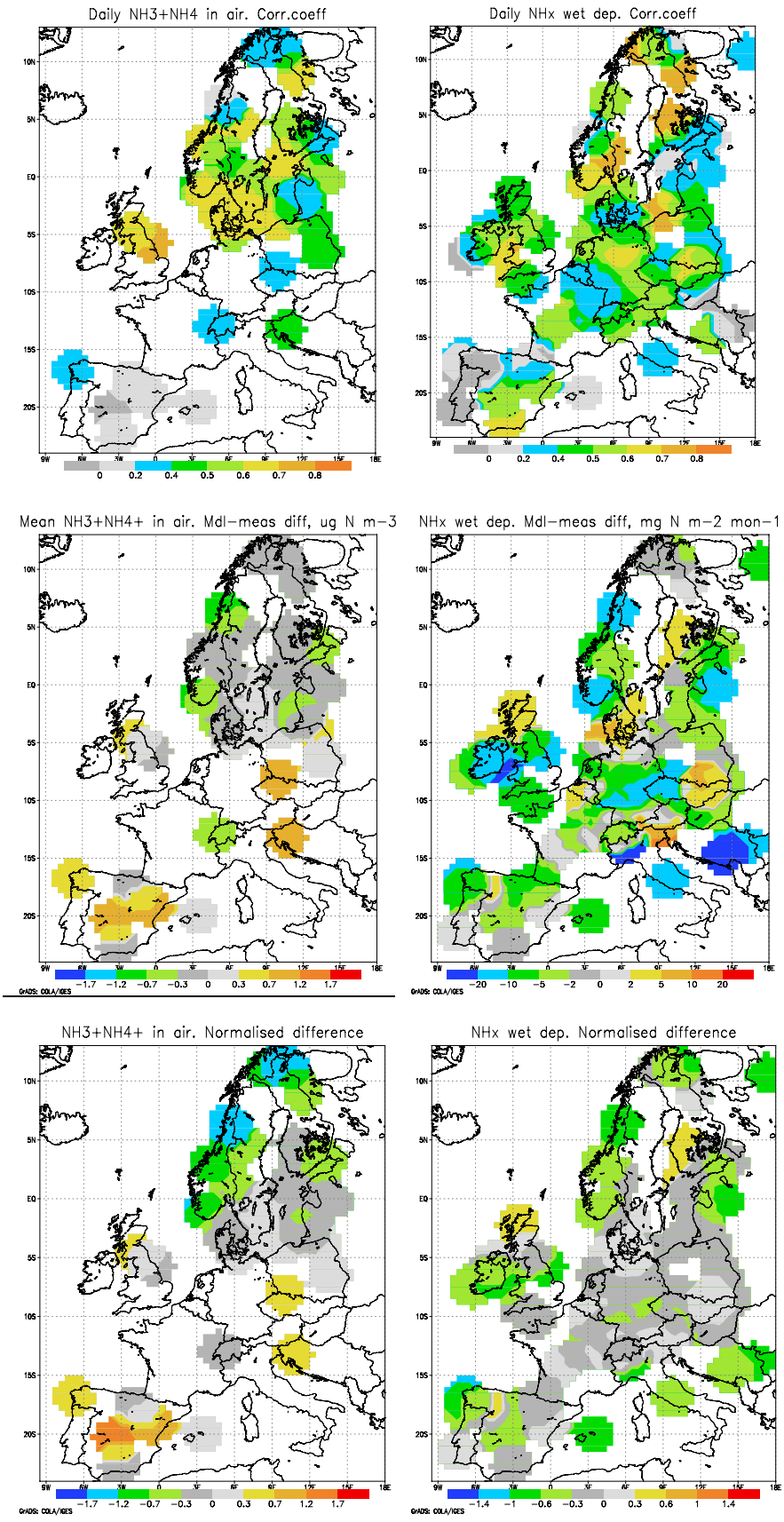


Fig. 7c: Upper panel: Spatial distribution of correlation between modelled and measured daily NH_3+NH_4 in air and monthly NHx wet deposition. Middle and lower panels: Spatial distribution of the absolute differences and the weighted absolute difference (unitless) of the same compounds.

The observed maximum wet deposition exceeding 100 mg(S) m^{-2} mon^{-1} in the Black Triangle region was overestimated up to ~160 mg(S) m^{-2} mon^{-1}. The model tendency to overestimate wet sulphate deposition continues for Central Europe, but usually does not exceed 30% of the mean value. For the rest of the domain, the absolute deviation is within ±15 mg(S) m^{-2} mon^{-1}. Usually, overestimation occurred during small-deposition events, while high-load episodes are underestimated. The correlation coefficients for monthly sums vary in a wide range from –0.1 for Portugal up to 0.8 for several Norwegian stations.

In the NMR-model domain, SO_2 was in general overestimated in winter at some south-western coastal stations and at one Swedish mountain station (Fig. 8). Underestimation occurs at some stations influenced by ship emissions and in northern stations influenced by the Kola Peninsula emissions. Monthly average standard deviations of daily values were at the same level or exceeded the monthly averages. The correlation is weak at some eastern and marine stations with low measurement frequency, and at clean and elevated sites, but was above 0.6 for most Finnish and south-western stations. The number of SO_4 aerosol samples was low due to the high detection limit of the instrument or other difficulties. Modelled SO_4-concentrations are underestimated at most stations, especially in summer.

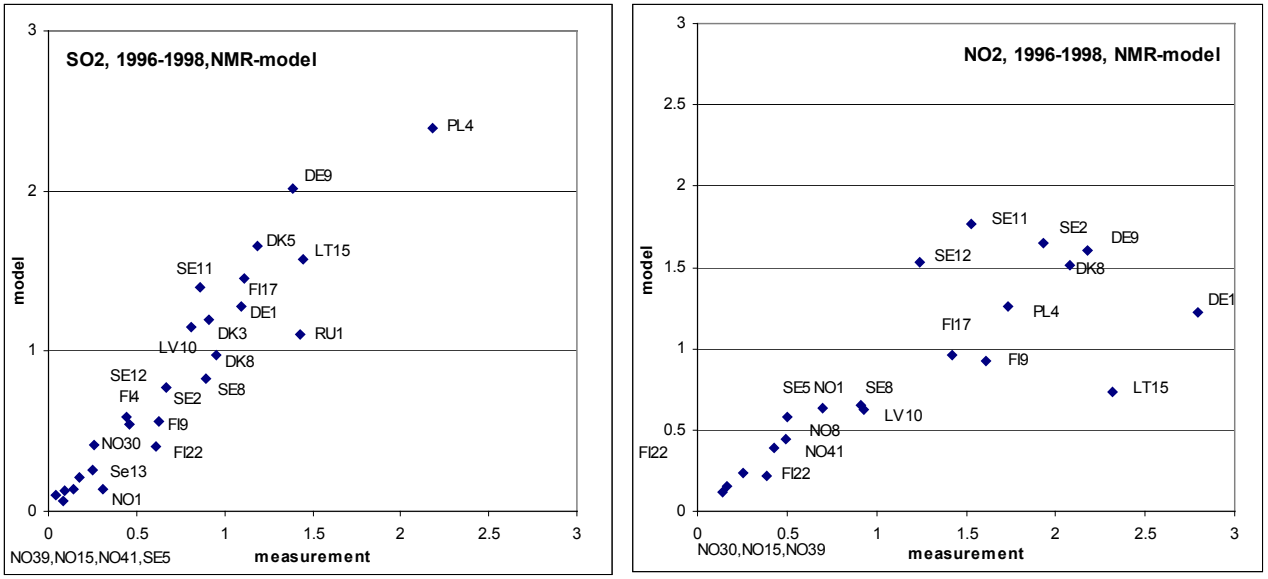


Fig. 8. Comparison of modelled and measured SO_2 and NO_2 mean concentrations. The NMR-model results over the period 1996-98.

The $SO_4^=$ wet deposition peaks were underestimated at most stations, while the modelled background deposition could be higher than the measured one. The underestimation is rather natural because, e.g., the non-European load is missing, and low precipitation was overestimated. The probability of rain is higher inside a grid-cell than at a single location. There was no difference between stations collecting weekly or daily samples in monthly results.

Reduced nitrogen

In the NMR-area, NH_3 was measured only in Latvia in 1997 and the Kola Peninsula in 96. It was underestimated especially in summer. NH_4^+ was slightly underestimated (~0.3 μg(N) m^{-3}) in the European domain by up to 25% in remote regions and less than 10% in Central Europe.

The sum NH_3+NH_4^+ was underestimated at remote Scandinavian sites, while in some parts of Europe the deviation had the opposite sign. The time correlation for NH_4^+ varies in a wide range with a maximum (0.5) over the southern Baltic region. The correlation of total ammonium is high over the whole Baltic Sea region (~0.6) and UK but statistically insignificant in the south.

Ammonium wet deposition is usually underestimated by 5-15 mg(N)m^{-2} mon^{-1} or about 20% in Central Europe. However, an overestimation of small-load events is clearly seen. Time correlation in the "good correspondence area" is about 0.5. There are also a few stations in France, Spain and the Black Triangle region where the agreement is very good. Over NMR-domain, the measured standard deviation and daily maximum NH_x wet deposition is much higher than the modelled, and no clear annual cycle in the deposition was detected. The correlation coefficients were generally fair. The monthly deposition peaks at Norwegian mountain stations, at the marine station of Hoburg and directly downwind of the large source areas were underestimated.

DISCUSSION AND CONCLUSIONS

A detailed analysis of the results shows that the model demonstrated quite good capabilities to describe both spatial and temporal characteristics of the sulphur and nitrogen pollution in the region surrounding the Baltic Sea. This "good correspondence area" covered also the British Isles, Benelux, North-western Poland, northern Germany and North-eastern France. The detailed stack emission inventory for Finland and its neighbouring eastern areas as well as the good calibration of the HIRLAM fields increase the NMR-domain model performance (Part I results). The southern part of the European domain is not so accurate. The 150 km resolution of the boundary conditions of the 1-layer EMEP-model for 1993-1995 also decreases the accuracy.

The episodic variation of air concentrations is much better reproduced during winters. The time correlation of practically all species between November and March exceeds the corresponding value for May-September by 0.1–0.2. The spatial patterns of agreement of mean concentrations over Europe are quite similar for both warm and cold periods. Mean levels of seasonal deposition are also quite coherent for both oxidised and reduced nitrogen deposition. The only exception is the sulphur load, where the above-noted general overestimation is specific only for the winter season. For summer, the model results are closer to the observed average deposition.

The rather coarse spatial resolution of the model and sometimes the high altitude of the station yield limited representativeness of the point observations in the model grid cells. The shorter averaging period of the data, the more severe the problem is. It affects all species, though in background areas, air concentrations have smoother patterns and thus these measurements can be extrapolated with less representativeness error. Wet deposition is largely determined by local precipitation, which at some stations, e.g., Utö in the Finnish archipelago, differs by 50% between two gauges, located by only 100 m from each other. At windy and mountainous stations, the extrapolation error for precipitation is large with daily and even monthly averaging. The overestimation of wet deposition was sometimes connected to the general overestimation of precipitation frequency and intensity.

Berg and Schaug (1994) showed that spatial representativeness is specific for each station, highly non-isotropic, non-stationary and depends on the particular substance. We can roughly estimate to 30% the relative standard deviation of daily concentrations and also monthly wet deposition. Consequently, the maximum possible time correlation coefficient between modelled and observed data is about ~0.7. Specific samples can exceed this level only occasionally. Uncertainties in the mean levels are smaller – they have to be attributed to the

systematic deviation of the station data caused, e.g., by a close emission source, specific local wind or relief patterns.

The scatter between modelled and measured SO_2, HNO_3+NO_3^-, sulphate and nitrate wet deposition are nearly the smallest possible in the "good-correspondence area" (determining the lower limit to the representativeness error) and moderate for NO_x, NH_x, and sulphate in aerosol. Such inconsistency between species in a single group (like sulphur or nitrogen compounds) leads to twofold conclusion. First, the core modules of the model like advection/diffusion, as well as related input data (e.g., 3-D wind) work well enough. On the other hand, substance-dependent parameterisation (either chemical transformation, or dry or wet sink rates) and corresponding input data (calculated OH and O_3 concentrations, boundary layer parameters and precipitation) have room for improvement. This conclusion is also supported by comparably high systematic deviations recorded in the absolute and relative differences and model-measurement regression for some species. There are systematic deviations even in good-fit areas that can be as large as 20-30% of the corresponding mean value.

Since grid-averaged concentrations in Eulerian models are obtained by mixing the individual plumes to the whole box volume and concentrations are further diluted through the advection algorithm, model values are lower than those detected. The underestimation of concentrations is also partly a consequence of zero non-European contribution and missing natural emissions.

In general, the Hilatar model showed good ability to describe the dynamical transport and dispersion of contaminants by atmospheric eddies, frontal patterns as well as in- and below-cloud scavenging on a scale of 25-50 km. Local or subgrid phenomena, such as sea breeze and seasonally changing coastal precipitation gradients, can be modelled only if the spatial resolution is below 10 km. Missing chemical sinks, such as photochemistry and chlorine reactions with nitric acid over sea areas, also increase uncertainty.

The study generated a data-base for environmental research projects containing the modelled fluxes and concentrations with 6-hour time resolution in a 26 km x 26 km over the Baltic Sea Area, and 0.5° grid over Europe. Baltic data starts from the year 1993, European data from June 1996, and the data base is continuously updated. Monthly deposition values up to the year 2000 will be available at www.fmi.fi, gridded numerical results can be requested from the FMI Air Quality Research Department.

Acknowledgements

We would like to thank the EMEP MSC-W Centre for their emission inventory and background concentrations for the years before 1996, Andreas Bott for his advection algorithm, the FMI HIRLAM group for the meteorological data and all FMI colleagues, especially Kirsti Jylhä, Ville Savolainen and Ilkka Vallinoja for working with the emission inventory or in common projects. Financial support from the EU BASYS project through MAST programme (contract: MAS3-CT96-0058), from the Nordic Council of Ministers and from the Academy of Finland is kindly acknowledged.

References

Berg T, & Schaug J, ed., 1994. EMEP workshop on the accuracy of measurements with WMO-sponsored sessions on Determining the Representativeness of measured parameter in a given grid square as compared to model calculations, Passau 1993. EMEP/CCC 2/94

EMEP 2000. Transboundary acidification and eutrophication in Europe. EMEP summary report CCC & MSC-W 2000. EMEP Report 1/2000, 264 p.

Hongisto, M., 1998 Hilatar, a regional scale grid model for the transport of sulphur and nitrogen compounds**.** Description of the model and simulation results for the year 1993. *Finnish Meteorol. Inst. Contributions No. 21*, Yliopistopaino, Helsinki, 152 p. presented in http://www.fmi.fi/research_air/air_25.html.

Hongisto, M., 2003. Hilatar, a regional scale multi-layer model for nitrogen and sulphur compounds over Europe and its Northern subregions. Part 1. Model Description. Part I in this journal.

Häkkinen, A.J., Markkanen, K., Hongisto, M., Kartastenpää, R., Milayaev, V.B., Yasensky, A., Kuznetsov, V.I., Kopp, I.Z. and Kivivasara, J., 1995. Emissions in St. Petersburg, the Leningrad area and Karelia. In: Tolvanen M., Anttila P., Kämäri J. (Eds). Proc. of the 10th World Clear Air Congress, Espoo, Finland, Vol 1, 103. The Finnish Air Pollution Prevention Society, 1995.

Leinonen L., (ed.), 1994-2001. Air Quality measurements 1993-2000. Official Statistics of Finland. Finnish Meteorological Institute, Helsinki. 250 p. each..

Lenhart, L., Heck, T. and Friedrich, R., 1997. The GENEMIS inventory, European emission data with high temporal and spatial resolution. In: *Ebel A., Friedrich R. and Rodhe H. (Eds). Transport and chemical transformation of pollutants in the troposphere Vol. 7. S*pringer-Verlag Berlin Heidelberg 1997, pp. 217-222.

Mäkelä, K. & Salo, M., 1994. Traffic emissions in Russia and the Baltic States. St. Petersburg, Leningrad Oblast, Republic of Karelia, Estonia, Latvia and Lithuania. *VTT Research notes 1569*.49 p.

Pietarila, H., Salmi, T. and Kivivasara, J., 1996. The Finnish National emission register. On-line documents, outputs and personal communication.

Ruoho-Airola T. and Salmi, T., 2001. Episodicity of sulphate deposition in Finland. Water, Air and Soil Pollution 130: 529-534.

Sofiev, M., 1999. Validation of model results on different scales. In *Approaches to scaling of trace gas fluxes in ecosystems*, A.F. Bouwman (Ed.). Developments in Atmospheric Science, 24, Elsevier, pp.235-255.

Vuorenmaa J., Juntto S. and Leinonen L., 2001. Rainwater quality and bulk deposition in Finland 1998. The Finnish Environment 468. Finnish Environmental Institute, Finnish Meteorological Institute 2001, 115 p.

Zlatev Z., Bergström R., Brandt J., Hongisto M., Jonson J.E., Lagner J. and Sofiev M., 2001. Studying sensitivity of air pollution levels caused by variations of different key parameters. TemaNord 2001:569. Nordic Council of Ministers, Copenhagen 2001. 47 p