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Comparison of modelled and monitored deposition fluxes of sulphur and nitrogen to ICP-forest sites in Europe

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Abstract

The EMEP MSC-W Eulerian chemical transport model, and its predictions of deposition of acidifying and eutrophying pollutants over Europe, play a key role in the development of emission control strategies for the UNECE and EU. It is important that this model is tested against observational data. Such model evaluation is usually conducted using observations from the EMEP monitoring network, which includes around 70 sites with wet deposition measurements in a variety of landscapes. Here we compare the results of the EMEP model with a completely independent data-set, 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 (r^2 values between 0.5–0.8 for most components and years, with mean values across all sites being within 30%), despite the uncertainty in comparing measured plot data with modelled grid data. EMEP tends to give somewhat lower values for the average, median and percentile SO_4^{2-}, NO_3^- and NH_4^+ 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 SO_4^{2-}, NO_3^- and NH_4^+ in precipitation are very similar as average (differences of 0–14%), median and percentiles, and the correlation between modelled and observed data is rather high (r^2=0.50–0.78). Many of the sites showing large discrepancies between EMEP 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. A simple precipitation variability index (PVI) was devised to flag sites with large variability, and many outliers in the chemical comparison were shown to be such sites. The largest discrepancies were seen between EMEP and ICP precipitation amounts and for this component the correlation was very poor (r^2=0.04–0.23 with all data, and 0.23–0.27 with PVI-filtered data). Although more work is needed to understand these discrepancies, the overall conclu-

sion is that the EMEP model performs rather well in reproducing patterns of S and N deposition to European forests.

1. Introduction

A major aim of EMEP (Cooperative Programme on the Long Range Transmission of Air Pollutants in Europe, http://www.emep.int) is to support governments with scientific guidance on the causes of air pollution concentrations and depositions within Europe. EMEP model results are an essential input to the RAINS integrated assessment model (Schöpp et al., 1999; Amann et al., 1999), and have been crucial to a number of United Nations Economic Commission for Europe (UNECE) Protocols and the recent European Union National Emissions Ceilings Directive. The modelling tool currently in use at the Meteorological Synthesizing Centre West (MSC-W) of EMEP is a Eulerian model which calculates concentration and depositions associated with acidification and eutrophication, as well as ozone (Simpson et al., 2003a). An essential part of the use of such a model has always been thorough evaluation against measurements, in order to give confidence in its basic scientific formulation and use for policy research. Typically, these evaluations have consisted of comparisons of modelled concentrations and deposition against measurements available though the EMEP Chemical Coordinating Centre (EMEP-CCC), which provides a network of 170 sites throughout Europe (Hjellbrekke, 2004). Seventy of these sites report wet deposition measurements.

The aims of the present work are twofold: (a) to compare the results of the EMEP model with a completely independent data-set, and (b) to compare EMEP simulations of deposition to forests with monitored deposition data from plots which were representative for forests in particular.

The data-sets used stem from the International Co-operative Programme on Assessment and Monitoring of Air Pollution Effects on Forests (ICP Forests, http://www.icp-forests.org). ICP forests was started in 1985, under the Convention on Long-Range Transboundary Air Pollution of UNECE. ICP Forests is responsible for the level

II monitoring system of forest sites, which has been in operation since 1994. The Forest Intensive Monitoring Co-ordinating Institute (FIMCI) was set up as a contractor of the European Commission (EC), to evaluate and validate these data (UNECE/EC, 2000). The monitoring data was collected from the EU/ICP-F Pan European Intensive (Level II) Monitoring Program via FIMCI (De Vries et al., 2001; ICP-Forest, 1998). The integrated monitoring of forest plots throughout Europe includes deposition monitoring, comprising throughfall, stemflow, bulk and wet only precipitation in open field sites (located close to the forest). This study includes comparisons of modelled and monitored data on an annual basis, as well as with seasonal resolution (monthly). This work was conducted as a cooperation between the the Swedish ASTA program (http://asta.ivl.se) and EMEP, and as part of the EU-funded NOFRETETE project (http://imk-ifu.fzk.de/nofretete/index.html).

The need for extensive model evaluation is strengthened by the complex nature of atmospheric deposition. An enormous number of complex and often poorly-understood processes, both physical and chemical, are involved in controlling the deposition of gases and particles from the atmosphere to vegetation (Fowler and Erisman, 2003; Erisman et al., 2005; Wesely and Hicks, 2000). Experimental evidence for processes such as possible compensation-points for NO or NH_3 (Duyzer and Fowler, 1994; Dorsey et al., 2004; Sutton et al., 1994), and co-deposition effects (Fowler et al., 2001) exists, but too few experiments are available to enable a reliable model approach.

Of course, comparisons of modelled versus observed concentrations have been published previously. Schaap et al. (2004) for example compared inorganic aerosol fields over Europe and found good agreement for mean levels (within 10–20%), and correlation coefficients of around r^2 ~0.27–0.38. Mathur and Dennis (2003) considered model performance for sulphate, nitrate and ammonium concentrations and depositions over the Eastern United States using a modified version of the RADM model (Chang et al., 1987). Model results for SO_4^{2-} and NO_3^- were found to be moderate to good (r^2=0.4–0.7), but much worse for NH_4^+. This work suggested severe problems with the NH_3 emission inventories. Similarly, Adams et al. (1999) have compared these compounds

for the globe, including some comparisons for EMEP measurement sites, obtaining good performance for SO_4^{2-}, with geometric means within 4% for the EMEP stations, degrading to within 63% for NH_4^+. In general, modelling of wet depositions is more difficult than for air concentrations. Model performance for wet deposition fluxes or concentrations cannot be better than the performance for meteorological data. Moreover, correct treatment of scavenging needs extensive access of meteorological parameters which are seldom available from standard meteorological drivers. Poor agreement between modelled and observed wet deposition fluxes has been seen both in a regional model intercomparison (van Loon et al., 2004) and for the global models in the COSAM model intercomparison (Roelofs et al., 2001).

Differences in model formulation, available emission inventories, and also in chemical climate between Europe and other parts of the World preclude the direct application of conclusions drawn from these studies to the EMEP model or to the situation over Europe. However, these results indicate strongly the uncertainties surrounding the modelling of the inorganic, and especially nitrogen-containing, components of the atmospheric aerosol. Further, since the EMEP model has special importance for the estimation of impacts of acidifying and eutrophying pollutants to European ecosystems, evaluation of this model's predicted deposition fields against measurements is an important component of building confidence in its use for both research studies and policy-oriented tasks.

2. Measurements

Total deposition (wet and dry) to forests can be estimated by measuring throughfall, including stemflow. The possibility to quantify the total deposition is limited to ions which do not take part in the uptake and leaching processes in the canopy, e.g. sulphate, sodium and possibly also chloride (Hultberg and Grennfelt, 1992; Erisman and Draaijers, 1995). If wet deposition (in the ICP network measured as bulk deposition in open field) is subtracted from measured throughfall flux, the remaining 'net' throughfall can

be used to estimate the dry deposition part to forests. Net throughfall includes not only the dry part, but also the possible contribution from fog and cloud water deposition.

Nitrogen in the form of both ammonium and nitrate, together with most other plant nutrients, is strongly affected by canopy exchange, especially in areas with low to moderate deposition of N. Different canopy exchange models have been proposed to separate between internal circulation and atmospheric deposition (Draaijers et al., 1996), but the uncertainties are relatively large (Erisman et al., 2005). For that reason modelled total deposition from throughfall data is not used in this study for comparison with EMEP modelled deposition of N to forests. Only measured bulk deposition of N in open field is compared with model calculated wet deposition by EMEP.

Quality-assured monitoring data during the period 1997 through 2000 was delivered by FIMCI, after approval from ICP Forest (De Vries et al., 2001; ICP-Forest, 1998). Data comprised bulk precipitation in open fields, throughfall in coniferous and deciduous forest plots and stemflow in some deciduous plots. As part of the FIMCI quality control, obviously contaminated or unrealistic values had been removed from the data set. Sites with data covering less than 315 days during a year were excluded from the study during that year.

However, some missing values (ca. 3% of the data) existed, which were filled-in by inserting the monthly mean value for that parameter, site and year to enable calculations of annual deposition. Following the above procedures, data from seven European countries (Sweden, France, Norway, Finland, Germany, Ireland, and Italy) representing various deposition levels, and with quality monitored deposition data were included in this study. This study includes 160 ICP sites in seven countries distributed over 140 EMEP grids (Fig. 1).

The ICP deposition to deciduous forests is uncertain due to the limited data on stemflow. Based on the stemflow data which was available (four sites in Germany and two sites in Sweden), factors to derive total deposition (including stemflow deposition) from throughfall deposition were derived. These factors were found to be 1.25 in Germany, and 1.1 in Sweden. The higher factor in Germany is due to a higher dry deposition, as

sources are located closer to the forest sites in Germany than in Sweden.

Inconsistent sampling periods complicate this comparison to some extent. ICP Forest data was collected at monthly intervals in Sweden and Finland, weekly in Ireland and parts of Germany, and on an irregular basis in Italy, Norway and some German counties. France collected data at 27 or 28 day intervals throughout the year (13 periods). Weekly or bi-weekly collections were transformed into monthly data if the break between resulting records fell within 4 days from the calendar months end. In accordance with ICP Forest guidelines, stemflow is only measured at level II sites with beech stands. However, data was not available for all the beech stands for which through-fall was provided. The available data of good quality was used to calculate a stemflow loss index to be used in estimating the total deposition to the forest floor as given by the EMEP model.

An important uncertainty in this comparison is associated with the precipitation sampling. A field inter-comparison of different bulk collectors used in the ICP network has found it difficult to estimate precipitation volumes accurately (Draaijers et al., 2001). The deviation for precipitation volumes compared to the best estimate ranged from +103% to −27%. Deviations are caused by several factors, such as aerodynamic properties and collecting area of the collector. It be noted that even when monitored by high-equality rain-gauges by official Meteorological Institutes, rainfall estimation is difficult. Smith and Fowler (2001) suggested that rainfall amounts for 5×5 km^2 in the UK could be uncertain by between 30%–50%.

Examination of the precipitation amounts reported for the ICP network shows some large differences between data for 1997 and 2000. The most extreme case involved precipitation of just 419 mm in 1997 but over 1200 mm in the year 2000. It is hard to judge how far such features can be ascribed to meteorological variability or to measurement artefacts (e.g. loss of precipitation in windy conditions, Nespor and Sevruk, 1999; Smith and Fowler, 2001), but some uncertainty should be attached to data where such year to year variability is large. In order to at least flag such sites, a precipitation

variability index, PVI, has been defined as percentage:

$$\mathrm{PVI} = 100 \times \left| 2. \frac{(P_{1997} - P_{2000})}{(P_{1997} + P_{2000})} \right| \quad (1)$$

where P represents the annual precipitation. Almost half the sites have a PVI of less than 10%, whereas 18% show PVI values of greater than 20%. Although all data-points have been used in all statistics in this paper, points with PVI values greater than 20% are indicated (circled) in scatter plots.

Finally, it should also be noted that although the ICP-Forest network forms an extensive and very useful data-set, there are limitations to the accuracy with which different components can be estimated. In an inter-comparison exercise of ICP samplers, Erisman et al. (2003) found that 65%, 50% and 45% of estimates for bulk precipitation of SO_4^{2-}, NO_3^- and NH_4^+ respectively, were more than 20% different from the best estimates. Further examples are discussed in Erisman et al. (2005).

3. The Eulerian EMEP model

For this study, regional concentrations of sulphur and nitrogen compounds have been calculated with the so-called OZONE version of the EMEP Unified Eulerian model, revision rv2_0. This model is fully documented in Simpson et al. (2003a) and Fagerli et al. (2004).

Briefly, the Eulerian EMEP model is a multi-layer atmospheric dispersion model for simulating the long-range transport of air pollution over several years. The model has 20 vertical layers in σ-coordinates and is primarily intended for use with a horizontal resolution of ca. 50×50 km^2 (at 60° N) in the EMEP polar stereographic grid. The chemical scheme uses about 140 reactions between 70 species, and makes use of the EQSAM module of Metzger et al. (2002a,b) to describe equilibria between the inorganic aerosol components.

All versions of the model use meteorological data from PARLAM (Benedictow, 2002), a dedicated version of the operational HIRLAM model (High Resolution Limited Area Model) maintained and verified at MET.NO. The anthropogenic emission input data used by the all model versions are generally based as far as possible upon emissions reported per sector and grid officially reported to the Convention on Long-Range Transboundary Air Pollution (e.g. Vestreng et al., 2004).

The dry deposition module makes use of a stomatal conductance algorithm which was originally developed for calculation of ozone fluxes, but which is now applied to all pollutants where stomatal control is important (Emberson et al., 2000a,b,c; Simpson et al., 2001, 2003b; Tuovinen et al., 2001, 2004). The deposition of SO_2 and NH_3 builds upon relationships presented in Smith et al. (2000) and Nemitz et al. (2001) involving temperature, humidity and the ratio SO_2/NH_3, in an attempt to allow for so-called co-deposition and surface acidity (Fowler and Erisman, 2003; Simpson et al., 2003a).

The model also allows for calculations to different types of land-cover within each grid. Dry deposition fluxes are calculated to a number of land-cover classes within each grid square, including two different classes of coniferous forest and two of deciduous forests. The resistance terms and deposition velocities are calculated independently over each land-cover, based upon vegetation characteristics such as height, leaf-area index, and phenology (Emberson et al., 2000b; Simpson et al., 2001). The greater height and associated roughness of forests leads to substantially greater deposition rates than are modelled over say grasslands or water surfaces.

Dry deposition of aerosol particles depends on their size, with the model version used here distinguishing between fine and coarse aerosols. Details of the formulation are given in Simpson et al. (2003a). Parameterisation of the wet deposition processes in the EMEP model includes both in-cloud and sub-cloud scavenging of gases and particles

The calculations presented here comprised monthly depositions for the years 1997 and 2000, separated in the forest classes coniferous and deciduous.

4. Results and discussion

The monitored plots with coniferous and deciduous forests were matched with the corresponding EMEP grid (50×50 km^2). Deposition data for SO_4^{2-} were compared for wet and total deposition separately. For NH_4^+ and NO_3^- only observations of wet deposition data could be used in the comparison for reasons discussed above.

Units used are given as kg (S or N) per hectare and year, as typically used in the deposition literature in Europe (1 kg ha^{-1} yr^{-1}=0.1 g m^{-2} yr^{-1}).

This study compares two completely different methods used to estimate deposition to forest locations: (a) measured data from a spot, normally a forest plot of around 30×30 m^2, with varying exposure, tree species, leaf area index, etc.; and (b) model calculated average deposition to the same forest type in a corresponding grid 50×50 km^2. It should be kept in mind that both methods have uncertainties and no true values are available. Of course, scatter is to be expected when paring single sites with grid data, but systematic differences between measurements at several monitored sites and model calculated values give valuable information on the causes and dimension of uncertainty. The comparison of modelled and observed data from 1997 and 2000 comprises:

1. precipitation amount,
2. total deposition of SO_4^{2-} to coniferous and deciduous forests,
3. wet deposition of SO_4^{2-}, NO_3^- and NH_4^+ in open field,
4. concentrations of SO_4^{2-}, NO_3^- and NH_4^+ in precipitation.

Total deposition from the ICP sites is represented by throughfall monitoring, representing both wet and dry deposition to forests, and wet deposition by bulk precipitation in open field. The results are compared to calculated wet and dry deposition from the EMEP grid model.

Results are shown as scatter plots and in tables of percentiles. Additionally Table 1 summarises the statistical parameters (intercept, slope and r^2 values) obtained with linear regression.

4.1. Precipitation

The precipitation amount at the ICP sites is normally measured in the same collectors that sample precipitation for chemical analysis and the methods are standardised in the ICP Forest manual. However, precipitation is notoriously difficult to measure accurately, and as noted in section 2, significant problems are known to affect ICP precipitation samples. Using the PVI index defined by Eq. (1) as an indicator of some of these problems, almost half the sites have a PVI of less than 10%, whereas 18% show PVI values of greater than 20%.

The comparison between precipitation used in the EMEP calculations and observed data at the ICP sites show large scatter (Fig. 2, Tables 1–2) with notably lower precipitation in EMEP grids expressed as average, median and percentiles, except for the areas with the lowest precipitation, the 20 percentile. The correlation coefficients (Table 1) are very low, although PVI-filtering makes a substantial difference to these values.

This can to some extent be explained by the coarse resolution of the model. The precipitation field pattern is very patchy (e.g. influenced by local topographic effects), and the regional scale model is unable to resolve this sub grid scale distribution. A typical problem arises with small scale showers. In reality precipitation is high in a small area of a given grid, but a large fraction of the grid should remain dry. Within the model, however, this precipitation is averaged out to cover the whole grid at a lower intensity. Thus, even though average precipitation amounts may be simulated well, the model predicts precipitation more often, but in lower amounts, than occur in reality. It can be noted, however, that the correlation between EMEP-model precipitation and measured precipitation was r^2=0.54 and r^2=0.42 for 1997 and 2000, respectively. This strongly suggests that some of the poor correlation is likely associated with ICP precipitation

methods.

4.2. Sulphur

Figure 3 compares the modelled (EMEP) and observed (ICP) deposition of SO_4^{2-} in coniferous forests for 1997 and 2000. The correlation is rather good during 1997 and 2000, especially if a few ICP sites with extremely high reported deposition values, or with PVI>20%, are excluded (Fig. 3 includes all points). This can also be seen from Fig. 4 where the modelled field of total sulphur deposition in 1997 has been plotted together with ICP data. This figure clearly shows a high degree of correlation between the EMEP and ICP estimates of total S deposition, with spatial gradients captured very well and no systematic errors across the sites.

Table 3 quantifies some statistics for this comparison. The results for 1997 are very good, with both mean and medium deposition values having errors of around 5%. For 2000 the error in the mean was low (4%), but much higher for the median (37%), with the EMEP model giving higher depositions in this case. This overestimate is unexpected, because the EMEP calculation represents an average forest with all age classes of trees. The ICP sites are normally located in mature forest stands, which generally reveal higher dry deposition than the most common forest stand.

The monthly resolution of modelled and observed SO_4^{2-} deposition was studied for 1997 with ICP sites in Germany (a high deposition area) and Sweden (moderate deposition), for which monthly deposition data was available (Fig. 5). The results for the Swedish sites are reasonable throughout the year. However, for Germany the EMEP results are significantly lower than ICP in February and December but significantly higher in September. It is unclear why the seasonal patterns should be so different for Germany, but one difference arises from the fact that the emission sources are located closer to the forest sites in Germany than in Sweden (forest composition is the same, coniferous). Deposition values are thus likely to more sensitive to local dispersion conditions in Germany than in Sweden, and the model may have some problems capturing this.

The modelled and observed total deposition of SO_4^{2-} in deciduous forests, shown in Fig. 6, show similar correlation as coniferous forests. As noted in section 2 above, the ICP deposition to deciduous forests is uncertain and correction factors were applied. With these corrections of the throughfall data, EMEP overestimated the SO_4^{2-} wet deposition slightly in most cases. The comparison between modelled and observed deposition of SO_4^{2-} in precipitation (Fig. 7) is more scattered compared to deposition in forests. EMEP is slightly underestimating the deposition as average, median and all percentiles, both 1997 and 2000 (Table 4), in comparison to observed data. The observed deposition in open field, collected with bulk samplers, could normally be expected to be 5 to 15% higher than wet only deposition, due to dry deposition in the open samplers in open field. However, the main reason for the higher deposition in observed data is higher measured precipitation amount at the ICP sites, in comparison to the values used in the EMEP calculations in the corresponding grids. The correlation of modelled and observed concentration of SO_4^{2-} in precipitation (Fig. 8) is higher and average, median and percentiles are very close (see Table 4), in comparison to deposition. The EMEP performance of modelling SO_4^{2-} deposition compared to observed data at ICP sites 1997 and 2000 can be summarised: EMEP slightly overestimates the total (wet and dry) deposition of SO_4^{2-} to forests, but differences are within 20% in 1997 and 30% in 2000, EMEP slightly underestimates the wet deposition of SO_4^{2-}, mainly due to lower precipitation amounts in the EMEP calculations. The modelled concentrations of SO_4^{2-} in precipitation is very similar to observed, but the modelled data should be 5 to 15% lower due to dry deposition in observed data in open field with bulk samplers. The consequence of the first two statements above is that EMEP seems to give somewhat higher dry deposition of SO_4^{2-} to forests, compared to the dry deposition observed in the ICP forest plots (throughfall minus deposition in open field).

4.3. Nitrogen Deposition

As noted in Sect. 2 it is only possible to compare modelled and observed nitrogen deposition in precipitation, because canopy exchange (uptake) of nitrogen affects the chemical composition of throughfall. Open-field sites are used for this comparison, and Fig. 9 compares modelled and observed deposition of NO_3^-. The results show that many points lie along the 1:1 line, but the scatter is large. However, many of the points lying far from the 1:1 line are also those associated with PVI values of more than 20%, which may indicate data-problems rather than model problems. The average, median and percentiles indicate an underestimation by EMEP, compared to observations (Table 5). The correlation between modelled and observed concentrations of NO_3^- is much higher (Fig. 10), compared to deposition, showing that the scattered picture in deposition is mainly caused by differences in precipitation amounts used by EMEP and ICP, and probably also by data problems at some ICP sites (indicated by high PVI values). The comparison between modelled and observed deposition of NH_4^+ (Fig. 13) in open field is very similar to deposition of NO_3^-, but the pattern is even more scattered for NH_4^+. The average and median values are at the same level for NH_4^+ (Table 6) and NO_3^- (Table 5). By comparing concentrations the correlation between modelled and observed data becomes much higher, except for high concentrations at a few ICP sites.

The spatial patterns of calculated and observed total depositions are illustrated in Figs. 11 and 12 for oxidised and reduced nitrogen respectively. These plots show mixed results, with values coinciding very well over most of France and northern Germany, but with some areas showing systematic differences between the EMEP and ICP deposition totals (e.g. southern Norway and Sweden).

The EMEP performance of modelling nitrogen deposition compared to observed data at ICP sites 1997 and 2000 can be summarised: EMEP slightly underestimates the average, median and percentile NO_3^- and NH_4^+ deposition in the whole deposition gradient, and the correlation between modelled and observed data is relatively low. Modelled and observed concentrations of both NO_3^- and NH_4^+ are very similar as av-

erage, median and percentiles, and the correlation between modelled and observed data is rather high. The differences in deposition are mainly caused by differences in precipitation amount used by EMEP and ICP.

4.4. Concentrations in precipitation

The differences in precipitation amounts used in EMEP and ICP are accompanied by a low correlation between modelled and observed data, as described earlier. The accuracy of the EMEP modelled wet deposition is probably better described by comparing modelled and observed concentrations in precipitation. The similarities between modelled and observed concentrations in precipitation are summarised in Table 7. The ICP sites in this study are probably too few to clearly show uncertainties in the data that EMEP uses, or to replace the precipitation data for the EMEP grids. But the observed systematic differences between EMEP and ICP data implies that the accuracy of the precipitation data used in EMEP should be evaluated against data from national precipitation networks or other independent data at the fine resolution that is required by the model.

4.5. Comparisons with EMEP sites

In order to put the above ICP comparison in the context, we present here a brief discussion of some relevant comparisons against wet deposition data from the standard EMEP network (CCC:2004), also for 1997 and 2000. The objective was to see whether the same conclusions on model performance could be drawn using the EMEP data as when using the ICP data. Such comparisons, albeit with earlier versions of the EMEP model have been presented in e.g. Fagerli et al. (2003); Fagerli (2004). Compared to these standard EMEP data, both oxidised and reduced nitrogen depositions are underestimated by the model in both years, with the same magnitude as in the comparison with ICP data. NO_3^- in precipitation is 15 and 23% lower than observations in 1997 and 2000, respectively, whilst NH_4^+ in precipitation is underestimated by approximately

10% in both years. However, in contrast to the comparison with ICP forest data, which concluded that the model underestimated sulphur wet deposition, model results for the EMEP sites slightly overestimate the measured data (Fig. 14). In 1997 and 2000 the average bias was 10 and 8%, respectively. The modelled precipitation is on average slightly higher than the amounts measured at the EMEP stations, also in contrast to the conclusions from the comparison with ICP precipitation data. There are no major differences in the overall model performance for the two years, although the wet deposition of oxidised nitrogen is somewhat more underestimated in 2000 than in 1997.

Further, as noted in Sect. 4.1, the correlation between EMEP-model precipitation and measured precipitation, r^2=0.54 and r^2=0.42 for 1997 and 2000 respectively, is much better than that found here between EMEP and ICP. This strongly suggests that some of the poor correlation is likely associated with the difficulties of sampling precipitation in the ICP network.

5. Conclusions

This paper has presented a comparison of the EMEP chemical transport model with a 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 sites of the EU/ICP Forest (Level II) monitoring network.

In general, similarities between modelled and observed deposition in this study were remarkably good, especially considering the many uncertainties in comparing measured plot data with modelled grid data, and uncertainties inherent in the deposition monitoring methods themselves (Erisman et al., 2005, 2003; Draaijers et al., 2001). It should be noted that the EMEP model, with it's grid size of approx. 50×50 km^2 (at 60° N), cannot be expected to reproduce small-scale variations in deposition regimes, caused by such factors as local emissions (especially important for NH_3, e.g. Sutton et al., 1998), topography (which has strong effects on rainfall amount and deposition,

e.g. Dore et al., 1992; Fowler et al., 1988), or where processes not included in the model (e.g. occult deposition) are important.

EMEP tends to give lower values for the average, median and percentile SO_4^{2-}, NO_3^- and NH_4^+ wet deposition in the whole deposition gradient compared to ICP, however, differences in mean values are within 20% in 1997 and 30% in 2000. Modelled and observed concentrations of SO_4^{2-}, NO_3^- and NH_4^+ in precipitation are very similar as average (differences of 0–14%), median and percentiles, and the correlation between modelled and observed data is rather high (r^2=0.50–0.78). The spatial pattern of S deposition was reproduced very well by the model. For oxidised and reduced nitrogen the overall spatial patterns were captured but systematic differences were found in some areas.

It was found that precipitation values reported by some ICP sites could show very large variation between 1997 and 2000. A simple variable, precipitation variability index (PVI) was defined to quantify this, and it was found that many of the sites showing large discrepancies between EMEP and ICP data were found to have unusually high PVI values. This suggests that some differences between EMEP modelled and ICP observed depositions may be due to sampling procedures or complex topographic effects. Although more work is needed to understand these discrepancies, the overall conclusion is that the EMEP model performs rather well in reproducing patterns of S and N deposition to European forests.

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Table 1. Summary of statistical comparisons for 1997 and 2000. Tables give intercept (c), slope (m), number of points (N) and correlation coefficients (r^2) derived from linear regression of the scatter plots indicated.

			All data				PVI < 20%			
Fig.[a]	Component[b]	Year	c	m	N	r^2	c	m	N	r^2
2	Precip.	1997	716	0.146	145	0.04	339	0.556	112	0.23
		2000	540	0.40	145	0.23	432	0.502	112	0.27
3	SO_4^{2-},CF	1997	1.97	0.779	112	0.74	2.83	0.737	85	0.71
		2000	3.64	0.516	112	0.45	3.02	0.688	85	0.64
6	SO_4^{2-},DF	1997	2.59	1.22	32	0.60	4.69	1.06	26	0.43
		2000	5.64	0.72	32	0.29	4.81	.856	26	0.37
7	SO_4^{2-},OF	1997	0.887	0.714	145	0.55	1.19	0.702	112	0.46
		2000	2.77	0.258	145	0.24	2.59	0.337	112	0.28
8	W-SO_4^{2-},OF	1997	0.0655	0.854	145	0.67	0.117	0.805	112	0.56
		2000	0.1125	0.672	145	0.54	0.112	0.632	112	0.50
9	NO_3^-,OF	1997	0.786	0.566	145	0.53	1.11	0.524	112	0.41
		2000	1.37	0.432	145	0.50	1.10	0.532	112	0.54
10	W-NO_3^-,OF	1997	0.045	0.754	145	0.76	0.073	0.717	112	0.69
		2000	0.035	0.792	145	0.79	0.044	0.785	112	0.78
13	NH_4^+,OF	1997	0.96	0.569	145	0.48	1.4	0.519	112	0.39
		2000	1.37	0.529	145	0.42	0.663	1.06	112	0.49

Notes: (a) Fig. refers to figure number in this paper; (b) prefix "W" stands for concentration in precipitation. DF for deciduous forest, CF for coniferous forest, and OF for open field. See relevant figure for more details.

Table 2. Annual precipitation during 1997 and 2000. EMEP modelled and ICP bulk (147 sites).

Annual precipitation	EMEP 1997 mm	ICP 1997 mm	EMEP 2000 mm	ICP 2000 mm
average	825	856	981	1105
median	719	823	896	1062
max	3038	2403	2865	2829
p80	917	1068	1126	1396
p60	794	898	951	1133
p40	691	729	841	932
p20	613	593	756	738
min	430	293	560	404

Table 3. Deposition of SO_4^{2-} (as S) in coniferous forests. EMEP modelled wet + dry and ICP throughfall (115 sites) during 1997 and 2000.

Deposition of SO_4^{2-}	EMEP 1997 kg/ha	ICP 1997 kg/ha	EMEP 2000 kg/ha	ICP 2000 kg/ha
average	8.30	7.90	7.31	7.00
median	5.96	5.63	6.81	4.98
max	32.31	38.34	19.11	30.78
p80	15.00	11.62	11.14	10.24
p60	7.73	7.36	7.51	6.57
p40	5.16	4.36	6.19	4.55
p20	2.60	3.04	3.45	2.94
min	0.59	0.83	0.78	1.06

Table 4. Deposition of SO_4^{2-} (as S) in precipitation. EMEP modelled wet and ICP bulk precipitation (149 sites) during 1997 and 2000.

Deposition of SO_4^{2-}	EMEP 1997 kg/ha	ICP 1997 kg/ha	EMEP 2000 kg/ha	ICP 2000 kg/ha
average	4.80	5.30	4.24	5.73
median	4.29	4.80	4.25	5.02
max	16.65	12.86	8.77	18.18
p80	6.96	7.83	5.33	7.93
p60	4.80	5.78	4.46	5.92
p40	3.60	4.29	4.04	4.40
p20	2.57	2.63	3.12	3.26
min	0.51	1.00	0.69	1.18

Table 5. Deposition of NO_3^- (as N) in open field. EMEP modelled wet and ICP bulk precipitation (157 sites) during 1997 and 2000.

Deposition of NO_3^-	EMEP 1997 kgN/ha	ICP 1997 kgN/ha	EMEP 2000 kgN/ha	ICP 2000 kgN/ha
average	3.07	3.65	3.28	4.44
median	2.92	3.37	3.30	4.16
max	6.56	8.26	6.50	12.41
p80	4.44	5.65	4.65	6.37
p60	3.34	4.09	3.88	4.74
p40	2.45	3.03	2.78	3.72
p20	1.66	1.74	2.05	2.13
min	0.30	0.36	0.39	0.34

Table 6. Deposition of NH_4^+ (as N) in open field. EMEP modelled wet and ICP bulk precipitation (157 sites) during 1997 and 2000.

Deposition of SO_4^{2-}-S	EMEP 1997 kg/ha	ICP 1997 kg/ha	EMEP 2000 kg/ha	ICP 2000 kg/ha
average	3.39	4.07	3.90	4.67
median	2.96	3.50	3.78	4.28
max	11.96	12.77	13.27	13.56
p80	5.38	6.31	5.82	7.16
p60	3.87	4.37	4.37	5.16
p40	2.26	2.84	2.94	3.63
p20	1.20	1.42	1.67	2.00
min	0.16	0.23	0.21	0.23

Table 7. Volume weighted concentrations in precipitation 1997 and 2000. Units given as mgS/L or mgN/L as appropriate.

Concentrations in precipitation		EMEP 1997 mg/L	ICP 1997 mg/L	EMEP 2000 mg/L	ICP 2000 mg/L
SO_4^{2-}	average	0.61	0.63	0.46	0.52
SO_4^{2-}	median	0.58	0.58	0.46	0.50
NO_3^-	average	0.39	0.45	0.36	0.41
NO_3^-	median	0.41	0.44	0.36	0.40
NH_4^+	average	0.42	0.49	0.42	0.43
NH_4^+	median	0.41	0.45	0.40	0.40

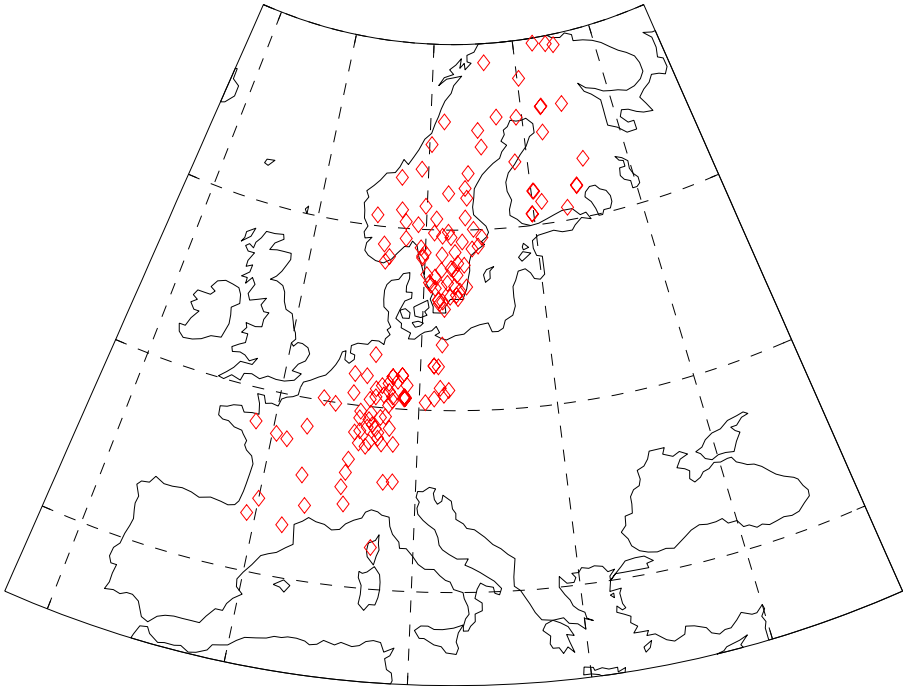

Fig. 1. Location of ICP sites used in this study.

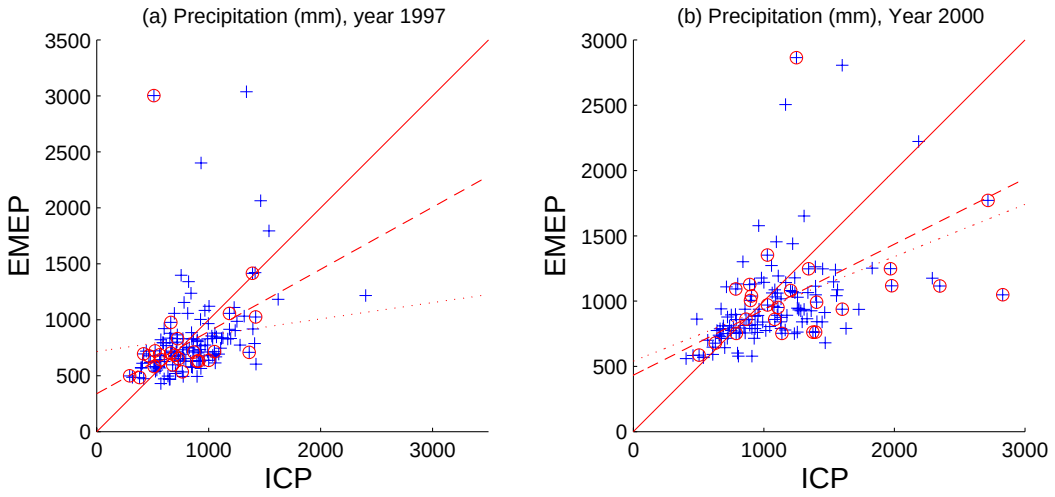

Fig. 2. Precipitation in mm during 1997 and 2000, EMEP modelled vs. ICP bulk. Circled points indicate sites with PVI>20%, see Sect. 2. Solid line represents 1:1 fit, dotted and dashed lines represent regression lines for all data, and for data where PVI<20%, respectively (see Table 1).

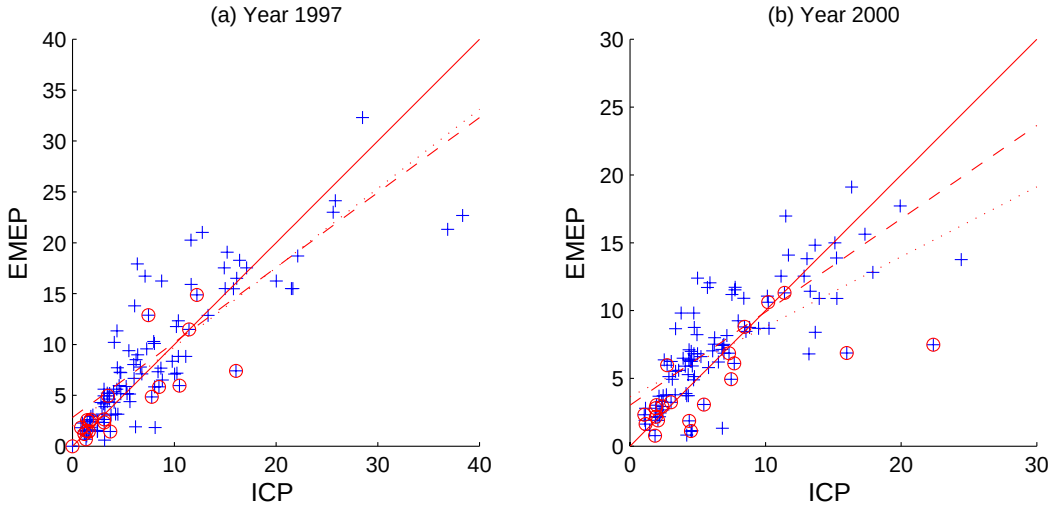

Fig. 3. Total deposition of SO_4^{2-} in coniferous forests, kgS ha^{-1} yr^{-1}. EMEP modelled wet + dry vs. ICP throughfall + stemflow during 1997 and 2000. Circled points indicate sites with PVI>20%, see Sect. 2. Solid line represents 1:1 fit, dotted and dashed lines represent regression lines for all data, and for data where PVI<20%, respectively (see Table 1).

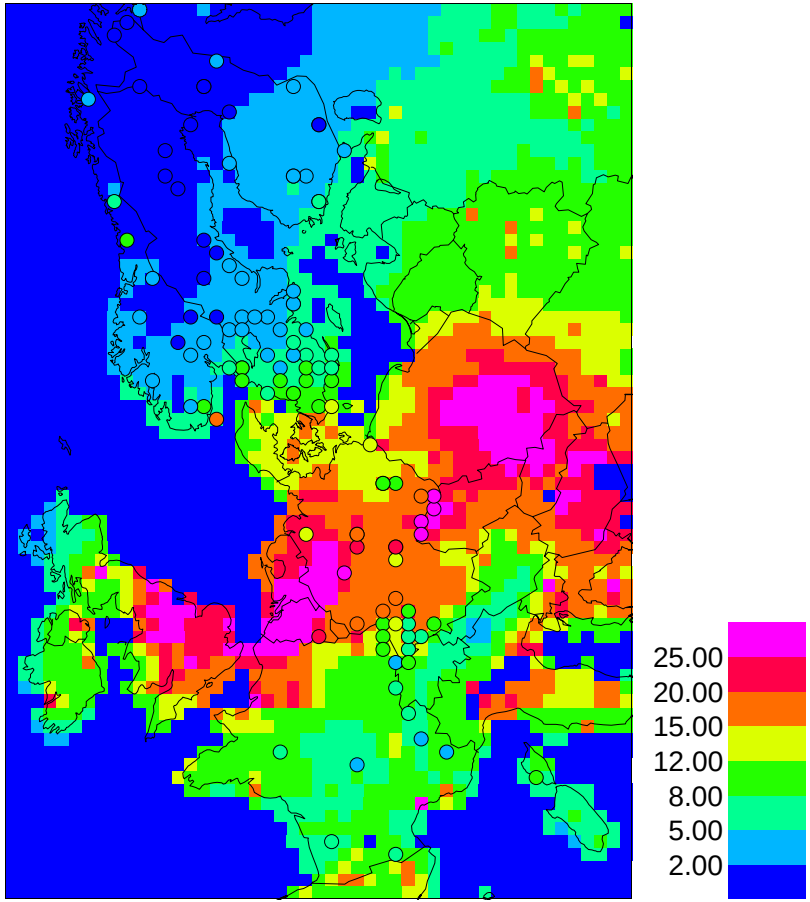

Fig. 4. Yearly average total deposition of SO_4^{2-} in coniferous forests, kgS ha^{-1} yr^{-1}. EMEP modelled wet + dry (deposition field) vs. ICP (bullets) throughfall during 1997.

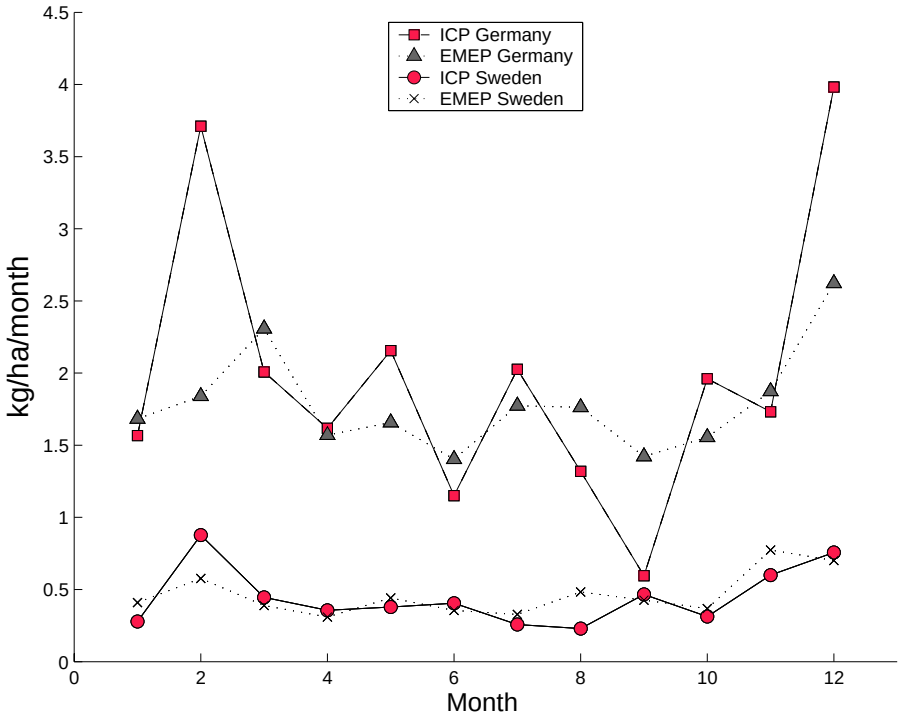

Fig. 5. Monthly average total deposition of SO_4^{2-} in coniferous forests, kgS ha^{-1} $month^{-1}$, 41 ICP sites in Sweden and 10 ICP sites in Germany. EMEP modelled wet + dry vs. ICP throughfall during 1997.

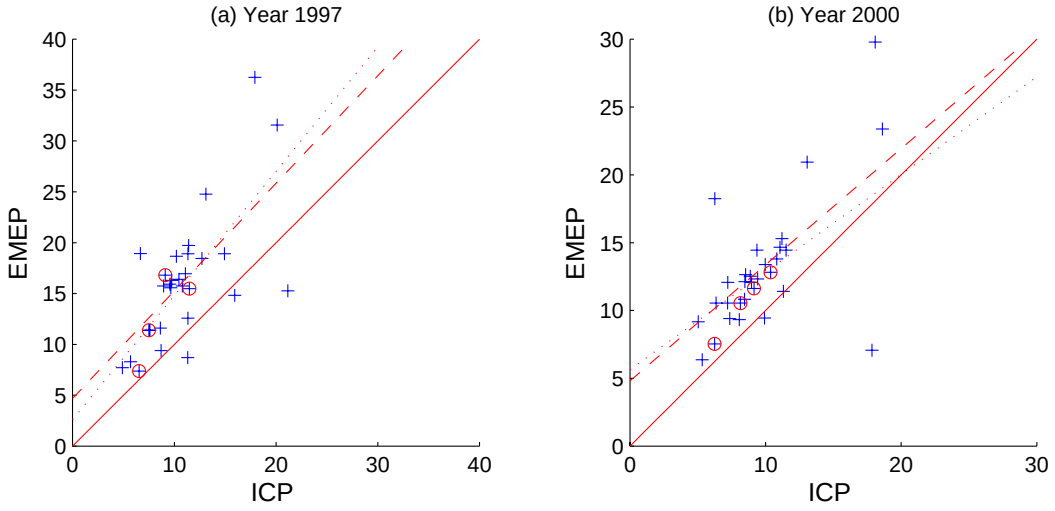

Fig. 6. Total deposition of SO_4^{2-} in deciduous forests, kgS ha^{-1} yr^{-1}. EMEP modelled wet + dry vs. ICP throughfall + stemflow during 1997 and 2000. Circled points indicate sites with PVI>20%, see Sect. 2. Solid line represents 1:1 fit, dotted and dashed lines represent regression lines for all data, and for data where PVI<20%, respectively (see Table 1).

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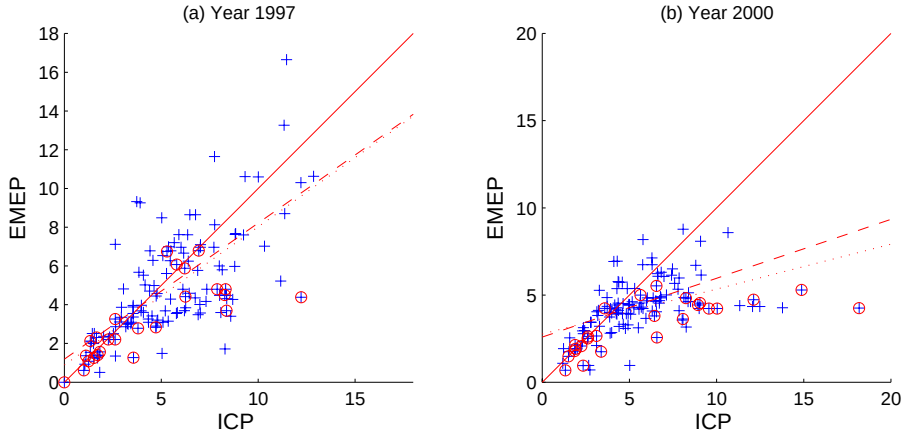

Fig. 7. Deposition of SO_4^{2-}-S in open field, kgS ha^{-1} yr^{-1}. EMEP modelled wet vs. ICP bulk precipitation during 1997 and 2000. Circled points indicate sites with PVI>20%, see Sect. 2. Solid line represents 1:1 fit, dotted and dashed lines represent regression lines for all data, and for data where PVI<20%, respectively (see Table 1).

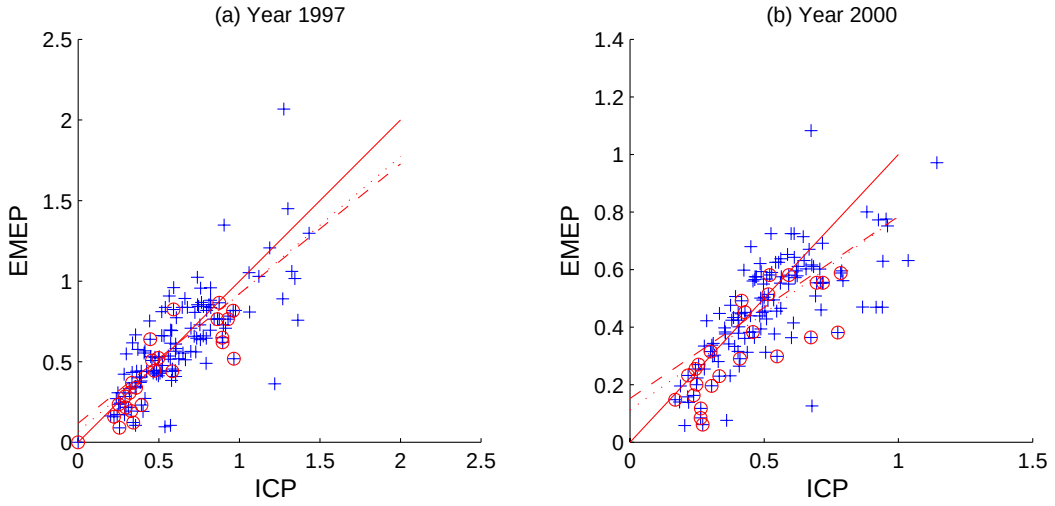

Fig. 8. Volume weighted concentrations of SO_4^{2-} in precipitation, mgS L^{-1}, EMEP modelled wet vs. ICP bulk precipitation during 1997 and 2000. Circled points indicate sites with PVI>20%, see Sect. 2. Solid line represents 1:1 fit, dotted and dashed lines represent regression lines for all data, and for data where PVI<20%, respectively (see Table 1).

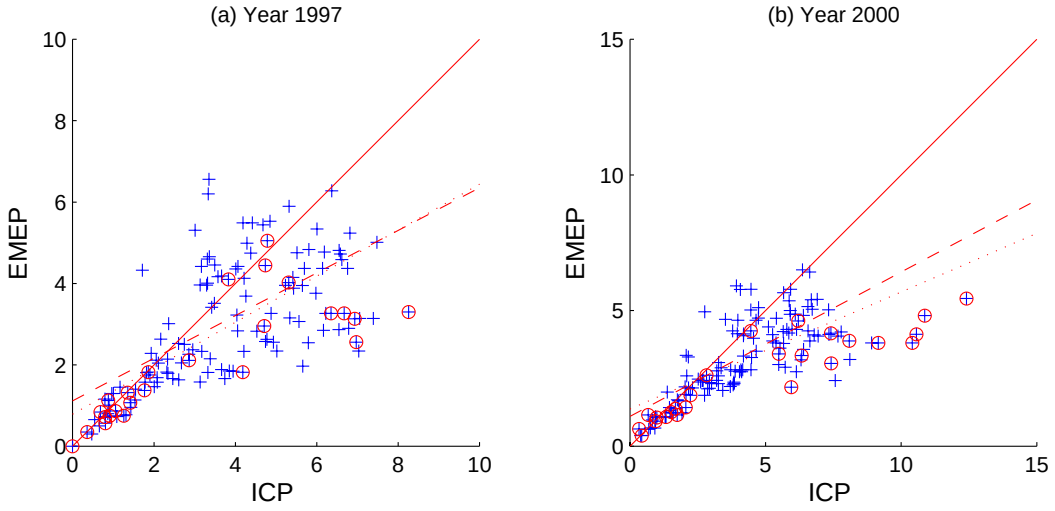

Fig. 9. Deposition of NO_3^- in open field, kgN ha^{-1} yr^{-1}. EMEP modelled wet vs. ICP bulk precipitation during 1997 and 2000. Circled points indicate sites with PVI>20%, see Sect. 2. Solid line represents 1:1 fit, dotted and dashed lines represent regression lines for all data, and for data where PVI<20%, respectively (see Table 1).

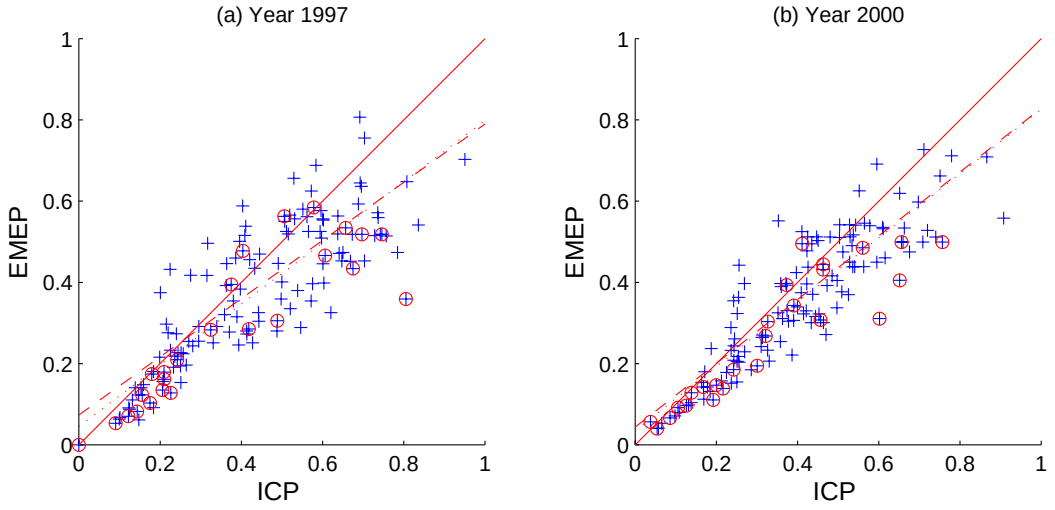

Fig. 10. Volume weighted concentrations of NO_3^- in precipitation, mgN L^{-1}, EMEP modelled wet vs. ICP bulk precipitation during 1997 and 2000. Circled points indicate sites with PVI>20%, see Sect. 2. Solid line represents 1:1 fit, dotted and dashed lines represent regression lines for all data, and for data where PVI<20%, respectively (see Table 1).

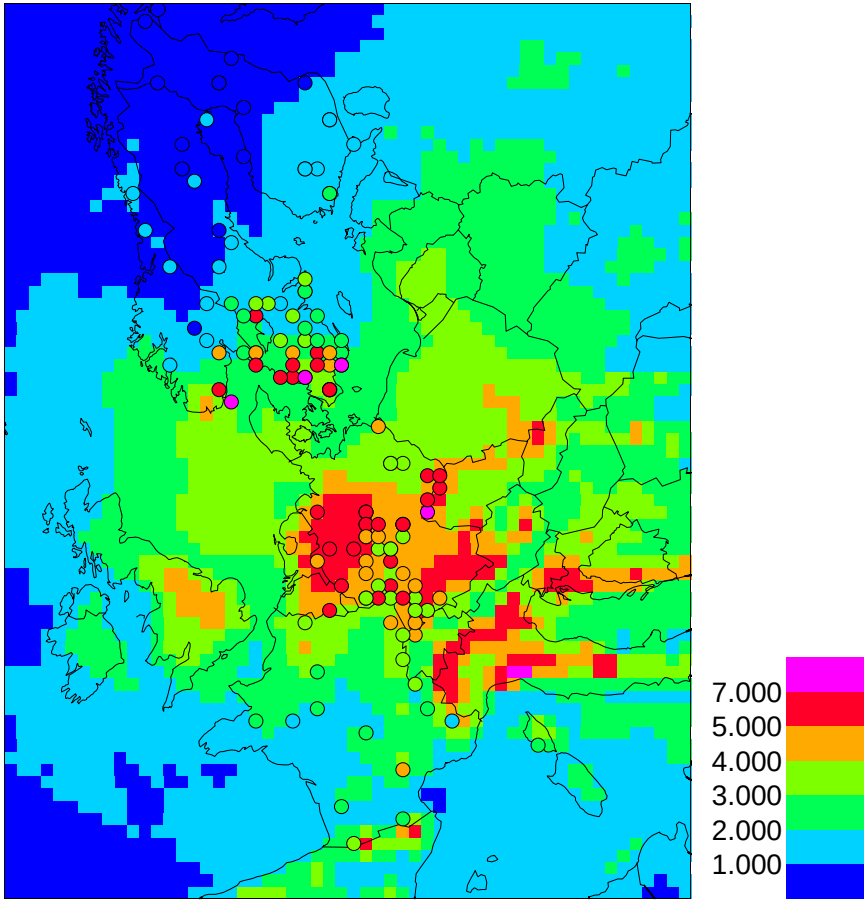

Fig. 11. Yearly average wet deposition of oxidised nitrogen, kgN ha^{-1} yr^{-1}. EMEP modelled wet deposition (deposition field) vs. ICP (bullets) during 1997.

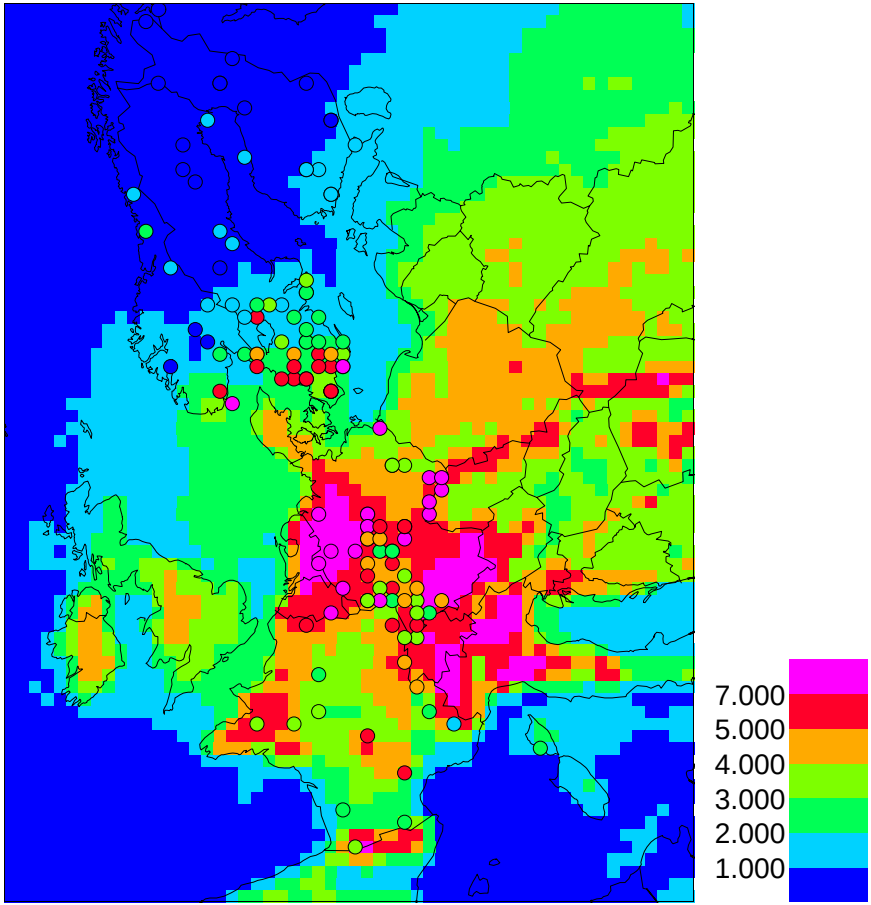

Fig. 12. Yearly average wet deposition of reduced nitrogen, kgN ha^{-1} yr^{-1}. EMEP modelled wet deposition (deposition field) vs. ICP (bullets) during 1997.

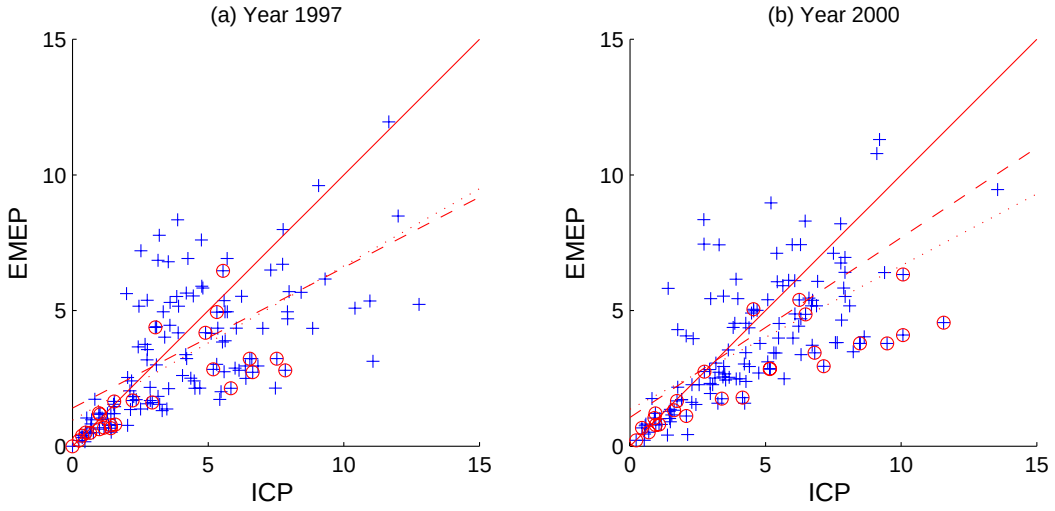

Fig. 13. Deposition of NH_4^+-N in open field, kgN ha^{-1} yr^{-1}. EMEP modelled wet vs. ICP bulk precipitation during 1997 and 2000. Circled points indicate sites with PVI>20%, see Sect. 2. Solid line represents 1:1 fit, dotted and dashed lines represent regression lines for all data, and for data where PVI<20%, respectively (see Table 1).

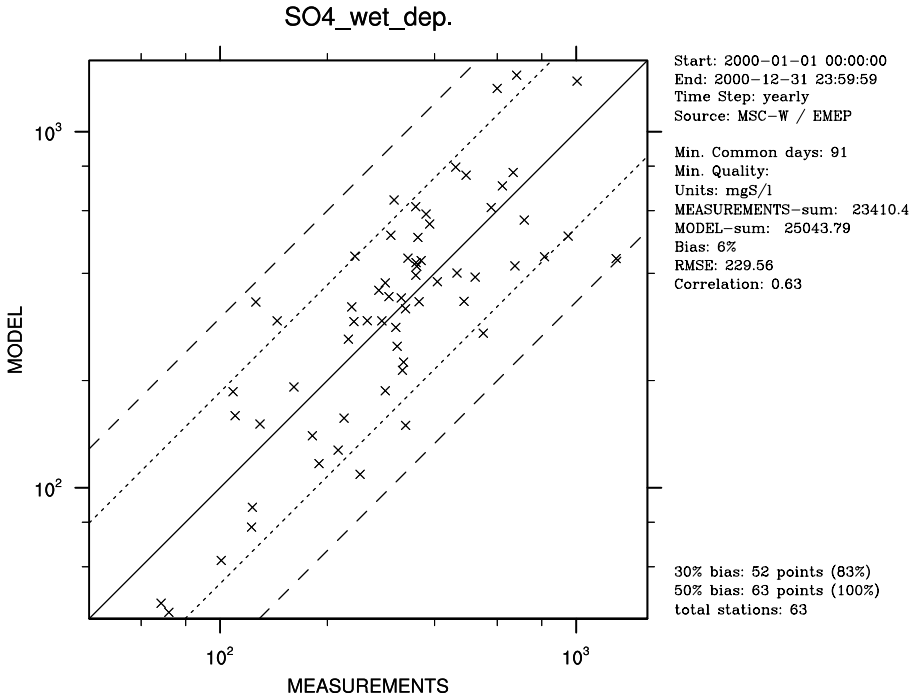

Fig. 14. Scatter-plots of modelled versus observed (EMEP stations) sulphur wet deposition in 2000 (μg(S)m^{-2}).

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