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**The RIVM-MNP contribution to the evaluation
of the EMEP Unified (Eulerian) model**

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Abstract

A few aspects of the EMEP Unified (Eulerian) model have been evaluated by analysing the deposition parameterisation for acidifying compounds and the concentration and deposition of SO_x, NO_x, and NH_x in the Netherlands. Evaluation was also carried out by analysing the source-receptor matrices for the Netherlands and the geographical distribution of the emissions, comparing results with both the OPS model and measurements. The picture given of the Netherlands by the EMEP Unified model was found for most acidifying compounds to be a fair one. The source-receptor matrices calculated by the EMEP and OPS models were seen to be in good agreement for oxidised sulphur, and in reasonable agreement for reduced nitrogen. Large discrepancies between the models were found for oxidised nitrogen. The contribution of the Dutch emissions to local deposition in the Netherlands came to a factor of 4 higher in the OPS model, compared to the EMEP model. The contributions of Belgium and Germany to deposition are also much higher in the OPS model. These differences can be traced back to the lower concentration and dry deposition, along with higher wet deposition, of NO_x in the EMEP model. For oxidised nitrogen, there was a large difference in the influence of the boundary and initial conditions on the source-receptor matrix. The EMEP model suggests that almost 30% of the deposition is due to sources outside Europe. The SO_2 concentrations in the Netherlands calculated with the EMEP model are close to the measurements, while the NO_x concentrations are about 40% lower and the NH_3 concentrations 30% to 40% lower than the measurements.

Preface

EMEP model output is used as input in the RAINS model, which, in turn, forms an important element of the scientific base for policy measures in the EU Clean Air for Europe (CAFE) programme and in the framework of the UNECE Convention on Long-Range Transboundary Air Pollution (CLRTAP). This report represents the results of evaluating the EMEP Unified model, as commissioned by the EMEP's Task Force on Measurements and Modelling (TFMM) through the EMEP Steering Body. The aim of the evaluation was to address the suitability of the model for assessing regional concentrations and long-range transboundary fluxes of sulphur, oxidised and reduced nitrogen, VOCs, ozone and suspended particulate matter. The activities described represent the RIVM-MNP contribution to the EMEP model's evaluation, intended to contribute to the quality of the EMEP model and the source-receptor matrices, and to assist in the policy-making process.

Contents

Executive Summary

A selected set of analyses has been performed by RIVM-MNP to evaluate the EMEP Unified (Eulerian) model. The analyses concentrated on the deposition parameterisation for acidifying compounds, the concentration and deposition of SO_x, NO_x, and NH_x in the Netherlands, the source-receptor matrices for the Netherlands and the geographical distribution of the emissions. For the evaluation, EMEP results were compared both with the OPS results and observations in the Netherlands to yield the following conclusions for the EMEP Unified model:

- **The EMEP Unified model was concluded to give a fair picture of the concentration and deposition of most acidifying compounds in the Netherlands.**

Source-receptor matrices

- **The source-receptor matrices for oxidised sulphur (SO_x) calculated by the EMEP and OPS models are in good agreement. The agreement for reduced nitrogen (NH_x) is reasonable.** For reduced nitrogen, the EMEP model calculated a higher contribution to the deposition in the Netherlands from emissions in Belgium and Germany, and a lower contribution from the Netherlands.

- **Large discrepancies between the EMEP and OPS models were found in the source-receptor matrices for oxidised nitrogen (NO_y).** The local deposition contribution of the Dutch emissions to the deposition in the Netherlands comes to a factor of 4 higher in the OPS model than in the EMEP model. The Dutch emissions contribute only 8% to the deposition in the Netherlands. This was 30-35% in the pervious version of the Eulerian model and 15-20% in the EMEP Lagrangian model. The NO_y contributions from Belgium and Germany are now also much lower in the EMEP model than in the OPS model. These differences may have consequences for emission reductions in countries to meet environmental objectives. When comparing the model's results with observations in the Netherlands, we see that the EMEP model strongly underestimates NO_x concentrations and dry deposition, and slightly overestimates wet deposition.

- **The relative contribution of Dutch emissions to the deposition in the Netherlands is always higher in the OPS model than in the EMEP model:** for SO_x (EMEP: 20%, OPS: 24%), NH_x (EMEP: 58%, OPS: 73%), and NO_y (EMEP: 8%, OPS 36%). This is in line with the findings that the OPS model calculates higher concentrations (NO_x) and/or uses higher dry deposition velocities.

- **There is a striking difference in the influence of the Boundary and Initial conditions on the source-receptor matrices for oxidised nitrogen.** The EMEP model suggests that about 30% of the deposition in the Netherlands is due to sources outside Europe, whereas in the case of oxidised sulphur, this is only 3%. This implies that for nitrogen oxide emission reductions, it might be necessary to go beyond Europe to reach environmental objectives for Europe.

Concentration and Deposition

- **The concentrations of SO_2 in the Netherlands as calculated by the EMEP model agree well with the measurements; however, the concentrations of NO_x and NH_3 show differences when compared with the measurements.** The SO_2 air concentrations in the Netherlands that were calculated with the EMEP model are similar to both the OPS results and the LML measurements. The NO_x concentrations calculated by the EMEP

model are about 50% lower than the OPS concentrations, and about 40% lower than the measured concentrations for all years. For the NH_3 concentrations, EMEP results are close to OPS results, but both are 30% to 40% lower than the measured concentrations, confirming the existence of the so-called 'ammonia gap'.

- **The wet deposition in the Netherlands as calculated with the EMEP model is about 25% higher for oxidised nitrogen than the measurements and about 20% lower for reduced nitrogen.** For oxidised sulphur the agreement with measurements improves, from about 30% higher for 1990 to about the same for 1997-2000. The wet deposition in the Netherlands as calculated by the EMEP model is about 20-50% higher for all compounds than the wet deposition calculated by the OPS model. The differences between the EMEP and OPS models for the wet deposition of oxidised sulphur decreased from 81% for 1985 to 19% for 2000; especially the last few years showed good agreement with the measurements.

- **The dry deposition in the Netherlands as calculated with the EMEP model is lower than that calculated with the OPS model.** On average, the EMEP model calculations for oxidised sulphur, oxidised nitrogen and reduced nitrogen were, respectively, about 15%, 55% and 35% lower than with the OPS model.

- **The total deposition (dry + wet) for oxidised sulphur and reduced nitrogen is about the same in both models.** This is due to higher wet and lower dry deposition in the EMEP model compared with the OPS model. The total deposition for oxidised nitrogen is about 20% lower in the EMEP than in the OPS model.

- **The parameterisation of dry deposition velocities in the EMEP model is not consistent.** The influence of temperature, humidity and surface wetness on the canopy resistance varies between NH_3 and SO_2. The (uncertain) effect of the co-deposition of SO_2-NH_3 was not modelled consistently for either gas .

- **It is recommended here to re-evaluate the dry deposition formulation of non-stomatal resistance, especially for wet conditions, on the basis of original experimental data.** Considerable discrepancies were found between models in the dry deposition of SO_x, NO_y, and NH_x, while the impact of high dry deposition velocities on SO_2 and NH_3 transport and deposition is large. Although the total (dry + wet) deposition of these compounds is modelled similarly in the EMEP and OPS models, the differences between the contributions of dry and wet deposition have large effects on source-receptor matrices, especially for NO_y.

- **The modelling of NO_x in the EMEP model is probably not correct; this has implications for the NO_x source-receptor matrices.**

Emissions

- **The geographical distribution of SO_x and NO_x EMEP emissions does not change in the course of 1980 to 2000.** This is valid for the low and high emission categories and all the sector emissions in a selected set of countries (Germany, The Netherlands, Poland), i.e., for all the years the same fraction of the total emissions of a sector is emitted per grid cell. This absence of changes in geographical distribution in emissions will likely affect the comparison between modelled and measured concentrations, especially for Eastern Europe.

- **The ratio between the EMEP emissions in the high and low emission categories is, per grid cell, the same throughout 1980-2000.** This holds for both the SO_x and NO_x emissions for all grid cells in the three selected countries.

Samenvatting

Een geselecteerde set analyses zijn uitgevoerd dor RIVM-MNP voor de evaluatie van het EMEP Unified (Euleriaanse) model. De analyses concentreerden zich op de depositie parameterisatie voor verzurende stoffen, de concentratie en depositie van SO_x, NO_x, en NH_x in Nederland, de bron-receptor matrices voor Nederland en de geografische verdeling van emissies. Voor de evaluatie zijn de resultaten van het EMEP model vergeleken met die van het OPS model en met metingen in Nederland. De volgende conclusies kunnen worden getrokken met betrekking tot het EMEP Unified model:

- **Het EMEP Unified model geeft vrij goede resultaten met betrekking tot de concentratie en depositie van de meeste verzurende stoffen in Nederland.**

Bron-receptor matrices

- **De bron-receptor matrices voor geoxideerd zwavel (SO_x) berekend door het EMEP model komen goed overeen met die van het OPS model. De overeenkomst voor gereduceerd stikstof (NH_x) is redelijk.** Voor gereduceerd stikstof berekend het EMEP model een grotere bijdrage aan de depositie op Nederland van België en Duitsland en een lagere bijdrage van Nederland.

- **Grote afwijkingen worden gevonden tussen het EMEP en OPS model voor de bron-receptor matrices voor geoxideerd stikstof (NO_y).** De lokale depositie bijdrage van Nederlandse emissies aan de depositie in Nederlandse is een factor vier hoger in het OPS model dan in het EMEP model. De Nederlandse emissies dragen slechts 8% bij aan de depositie in Nederland. In de vorige versie van het Euleriaanse model was dit 30-35% en in het EMEP Lagrangiaanse model 15-20%. De NO_y bijdragen van België en Duitsland zijn nu ook veel kleiner in het EMEP model dan in het OPS model. Deze verschillen kunnen gevolgen hebben voor de emissiereducties in landen welke nodig zijn om milieukwaliteitsdoelstellingen te halen. Een vergelijk van de resultaten met metingen in Nederland laat zien dat het EMEP model de NO_x concentraties en droge depositie sterk onderschat en de natte depositie licht onderschat.

- **De relatieve bijdrage van de Nederlandse emissies aan de depositie in Nederland is altijd hoger in het OPS model dan in het EMEP model.** Voor SO_x (EMEP: 20%, OPS: 24%), NH_x (EMEP: 58%, OPS: 73%) en NO_y (EMEP: 8%, OPS 36%). Dit is in overeenstemming met de bevindingen dat het OPS model hogere concentraties (NO_x) berekend en/of hogere droge depositie snelheden gebruikt.

- **Er is een opvallend verschil in de invloed van de Grens- en Begincondities op de bron-receptor matrices van geoxideerd stikstof.** Het EMEP suggereert dat ongeveer 30% van de depositie in Nederland is afkomstig van bronnen buiten Europa, terwijl dat slechts 3% is voor geoxideerd zwavel. Dit dat er emissies reducties voor stikstofoxiden buiten Europa nodig om milieukwaliteitsdoelstelling in Europa te halen.

Concentratie en depositie

- **De door het EMEP model berekende concentratie van SO_2 in Nederland komt goed overeen met metingen, maar de concentraties van NO_x en NH_3 vertonen verschillen ten opzichte van de metingen.** De concentraties in de lucht in Nederland berekend met het EMEP model liggen dicht bij de resultaten van het OPS model en dicht bij de LML metingen. De NO_x concentraties van het EMEP model liggen ongeveer 50% lager dan de OPS concentraties en ongeveer 40% lager dan de metingen voor alle jaren. Voor NH_3

liggen de EMEP resultaten dicht bij de OPS resultaten, maar beide zijn 30% tot 40% lager dan de metingen, waarmee beide modellen het bestaan van het zo genoemde 'ammoniakgat' bevestigen.

- **De natte depositie in Nederland berekend met het EMEP model ligt voor geoxideerd stikstof ongeveer 25% hoger dan de metingen en voor gereduceerd stikstof ongeveer 20% lager.** De overeenkomst met de metingen voor geoxideerd zwavel verbetert van ongeveer 30% hoger in 1990 tot ongeveer gelijk in 1997-2000. De natte depositie in Nederland berekend met het EMEP model is voor alle stoffen ongeveer 20-50% hoger dan berekend met het OPS model. Het verschil tussen beide modellen in natte depositie van geoxideerd zwavel neemt af van 81% in 1985 tot 19% in 2000 en speciaal voor de latere jaren is er een goede overeenkomst met de metingen.

- **De droge depositie in Nederland berekend met het EMEP model is lager dan berekend met het OPS model.** Gemiddeld is het EMEP model ongeveer 15%, 55% en 35% lager dan het OPS model voor respectievelijk, geoxideerd zwavel, geoxideerd stikstof en gereduceerd stikstof.

- **De totale depositie (droog + nat) is ongeveer gelijk in beide modellen voor geoxideerd zwavel en gereduceerd stikstof.** Dit komst door de hogere natte en lagere droge depositie in het EMEP model ten opzichte van het OPS model. De totale depositie voor geoxideerd stikstof is ongeveer 20% lager in het EMEP dan het OPS model.

- **De parameterisatie van de droge depositiesnelheden in het EMEP model is niet erg consistent.** De invloed van de temperatuur, vochtigheid en oppervlakte natheid op de canopy weerstand wordt verschillend behandeld voor NH_3 en SO_2. Tevens wordt het (onzekere) effect van de co-depositie SO_2-NH_3 niet consistent gemodelleerd voor beide gassen.

- **Het wordt aanbevolen om de formulering van de droge depositie voor de niet-stomatale weerstand, speciaal voor natte condities, te herevalueren aan de hand van de originele experimenten.** Er zijn aanzienlijke verschillen tussen modellen in de droge depositie van SO_x, NO_y en NH_x, terwijl de invloed van grote droge depositiesnelheden op het transport van SO_2 en NH_3 groot is. Alhoewel de totale (droog + nat) depositie van deze stoffen ongeveer hetzelfde is voor het EMEP en OPS model, hebben de verschillen in de bijdragen van de droge en natte depositie grote effecten voor de bron-receptor matrices, speciaal voor NO_y.

- **De modellering van NO_x in het EMEP model is waarschijnlijk niet correct, hetgeen implicaties heeft voor de bron-receptor matrices.**

Emissies

- **De geografische verdeling van de SO_x en NO_x EMEP emissies blijft constant door de jaren heen in de periode 1980, 1985-2000.** Dit geldt zowel voor de hoge en lage emissiecategorieën als voor alle sectoremissies in een geselecteerde set landen (Duitsland, Nederland, Polen), dat wil zeggen, voor alle jaren wordt per gridcel dezelfde fractie van de totale emissies van een sector geëmitteerd. Dit ontbreken van een verandering in de geografische verdeling van de emissies kan effecten hebben voor het vergelijk tussen gemodelleerde en gemeten concentraties, speciaal voor Oost Europa.

- De verhouding tussen de EMEP emissies in de hoge en lage emissiecategorieën is, per gridcel, hetzelfde voor alle jaren in de periode 1980, 1985-2000. Dit geldt voor alle gridcellen in de drie geselecteerde landen en voor de SO_x en NO_x emissies.

1.   Introduction

The EMEP Steering Body commissioned its Task Force on Measurements and Modelling (TFMM) to arrange for an evaluation of the new EMEP Unified (Eulerian) model as part of its working plan for 2003. At its 4th meeting in Valencia, Spain, in April 2003, the Task Force agreed on the general terms of the review to address the suitability of the model for assessing the regional concentrations and long range transboundary fluxes of sulphur, oxidised and reduced nitrogen, VOCs, ozone and suspended particulate matter. The Task Force agreed to have the review, commissioned by the Steering Body, completed for a workshop in Oslo in November 2003.

The EMEP Eulerian Model evaluation consists of three elements:

I.   Examination of processes included in the EMEP Eulerian model.

II.   Evaluation of the model performance compared with observations of key species.

III. Evaluation of the source-receptor relationships for sulphur, nitrogen, ozone and suspended particulate matter (PM mass).

Contributions from experts participating in the work of the TFMM are expected to:

* Consider the choice of process and meteorological parameterisations, chemical mechanisms and the sources of model input data employed in the EMEP Model.

* Quantify what would be considered as state-of-the-art in terms of model performance compared with observations based on their own national modelling studies.

* Identify key field campaigns for model evaluation purposes.

* Examine source-receptor relationships determined from their own national modelling studies.

More details on the items I-III and the possible contributions from experts can be found in a document by MSC-West (L. Tarrasón). The results of the Eulerian model evaluation can be found in three EMEP reports (Simpson, 2003; Tarrasón, 2003a,b).

TNO-MEP and RIVM-MNP have co-ordinated their contributions to the evaluation. The TNO-MEP contribution consists of a model intercomparison aimed at establishing the performance of the EMEP model compared with other models for several years (in task II-5) and of scenario calculations with the LOTOS model (in task III-3).

The RIVM-MNP contribution, described in this report, consisted of the following:

* Evaluation of the EMEP deposition parameterisation for acidifying compounds (in task I-3); see section 2. Documentation on the parameterisations used in the EMEP model is compared with information available in the literature. Deposition velocities of primary (SO_2, NO_x, NH_3) compounds are compared with values in the RIVM-MNP's DEPAC deposition parameterisation scheme (also used in the OPS model).

* Evaluation of EMEP model results on a finer grid scale (in task II-6); see section 3. The grid size of the EMEP model has decreased from 150 x 150 km to a current resolution of 50 x 50 km. The locations of the EMEP observational network used to validate the EMEP model are located in Europe on background sites that are widely separated. The performance of the EMEP model on the 50 x 50 km grid is tested by comparing the EMEP results with output from the OPS model (5 x 5 km grid) for yearly average

concentrations and depositions of SO_2, NO_x, and NH_3 for the Netherlands for 1985–2000. In addition, the results of both models are compared with spatially averaged observations of the RIVM's Netherlands air quality monitoring network (LML).

- Evaluation of source-receptor relationship for the Netherlands (in task III); see section 4. The EMEP source-receptor relationship (blame-matrices) for NO_x, SO_x and NH_x depositions are compared with the OPS results, with the focus on the Netherlands, i.e. emissions in the Netherlands compared to deposition there, and foreign emissions compared to deposition in Netherlands for the year 2000.

- Evaluation of the geographical distribution of emissions used by the EMEP model (in task I-3); see section 5. The changes in geographical distribution of the SO_x and NO_x emissions of several countries is compared for 1980-2000.

This evaluation is clearly not comprehensive. Due to the strict time-frame for the whole EMEP evaluation (less than six months) and the limited available time at RIVM-MNP for this new project (10 weeks), we had to make use of available data and analyses. The analyses discussed in this report are therefore limited to statements and observations.

The research described in this report was presented at the EMEP's TFMM meeting in Oslo on November 3-5, 2003.

2.　Deposition parameterisation

The dry deposition process is an important removal process for almost all gas-phase atmospheric pollutants. In emission areas it even dominates over wet removal processes, certainly if emission sources are close to the surface. As such, it largely determines not only the long-range transport of pollutants, but also the actual deposition load to ecosystems. The results of modelling wet deposition can be verified with (direct) observations. This is, unfortunately, not the case with dry deposition and, therefore, a comparison between models can be considered as the next best approach.

Dry deposition is usually described in transport models with a dry deposition velocity, where the dry deposition load is the product of the dry deposition velocity (V_d) and the atmospheric concentration. In the first long-range transport models, a fixed velocity was used, e.g. 0.008 m s^{-1} for SO_2. Such velocities were usually based on literature reviews of field experiments. In a next stage V_d was simulated with the so-called resistance model, in which (substance-independent) meteorological processes were distinguished from and substance-specific processes, the former usually expressed as the aerodynamic resistance and the latter as the canopy resistance. In the present Eulerian EMEP model an attempt has been made to split the substance-specific processes into sub-processes such as stomatal leaf uptake and non-stomatal leaf surface uptake, the latter in relation to the level of other pollutants (co-deposition).

Dry deposition velocities can be measured in the field. The most common method is to determine the dry deposition-induced vertical concentration gradients. In practice this means that very small concentration differences have to be measured, which requires extremely sensitive and stable equipment. In addition, one has to be extremely careful to avoid local disturbances. In principle, the outcome of such experiments is only valid for a specific substance, a specific vegetation type and (probably) a specific pollution climate. It is not surprising that reviews of the earlier publications of dry deposition velocities showed a very wide range of values.

Next to field campaigns one can simulate (measure) the dry deposition process in a laboratory. The advantage is that the conditions can be controlled and the influence of sub processes can be examined, but the disadvantage is that it is hard to translate laboratory results to field circumstances.

In the present context it is not so much the question if certain dry deposition processes (such as co-deposition) exist, but more the question of there being enough data to support practical parameterisations. Several years ago, the dry deposition parameterisation scheme, DEPAC, (Erisman *et al.*, 1994) was developed at RIVM. This scheme is based on measurements carried out in the framework of the EUROTRAC/BIATEX programme and on literature data. DEPAC has been implemented in (inferential) models such as EDACS (Van Pul *et al.*, 1995; Erisman and Draaijers, 1995), in the Dutch OPS model (Van Jaarsveld, 1995) and in (an older version of) the EMEP model (Seland *et al.*, 1995). Since then experimental work has been done in the EC-financed LIFE programme (Erisman *et al.*, 2001) and in the framework of EUROTRAC-2 sub-project, BIATEX-2 (GRAMINAE project, Sutton *et al.*, 1999).

2.1 Comparison of overall dry deposition velocities EMEP and OPS

Nowadays dry deposition parameterisations have so many variables that it not easy to see what the relevant differences are. Therefore, as a first comparison, the effective annual average dry deposition velocity is calculated for the EMEP and OPS models. This is done by dividing the total dry deposition flux by the concentration of the primary substance (SO_2, NO_x, NH_3). This does not exactly provide the dry deposition velocity for the primary substance, since total dry deposition includes the dry deposition of secondary products. However, in case of the Netherlands, the error made is not large and, moreover, it is for purposes of comparison only. The effective dry deposition velocities are shown for the EMEP and OPS models in Figure 2.2.

The first conclusion is that year-to-year variations are quite similar, probably pointing to meteorological influences being similar in both models. The dry deposition velocities in the OPS model are remarkably higher than in the EMEP model for SO_2 and NH_3, and comparable for NO_y. To see what causes the difference between the two models it is necessary to look at the parameters used in both models.

The resistance model for dry deposition of SO_2 or NH_3 to vegetation, applied in the EMEP model, is given in Figure 2.1. Common in almost all models are the aerodynamic resistance R_a and the quasi-laminar layer resistance R_b. Especially R_a depends strongly on the roughness of the surface and is therefore very different for such aspects as forest- and grass-covered surfaces. Dry deposition is assumed to take place along two pathways: via the stomata characterised by R_{stom} and via other (external or non-stomatal) pathways, here characterised by a single resistance R_{ns}. Of these two resistances, R_{stom} is the most elaborated in the literature and has, most of the time, by far also the highest value of the two. Therefore we concentrate on R_{ns}. This resistance represents the uptake of SO_2 and NH_3 on the external parts of the vegetation, e.g., in droplets or water films at the leaf surfaces. It is therefore a function of the chemical composition of the moisture, temperature, air humidity and/or surface wetness.
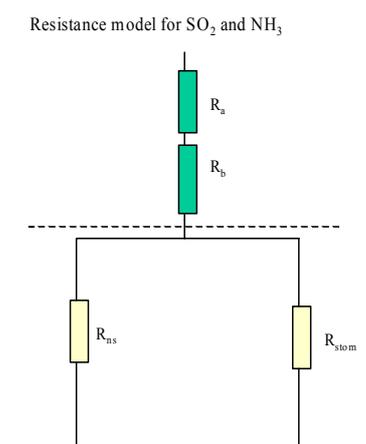
Figure 2.1. The resistance model applied in the EMEP model.

Effective dry deposition velocity SO2;

Effective dry deposition velocity NOx;

Effective dry deposition velocity NH3;
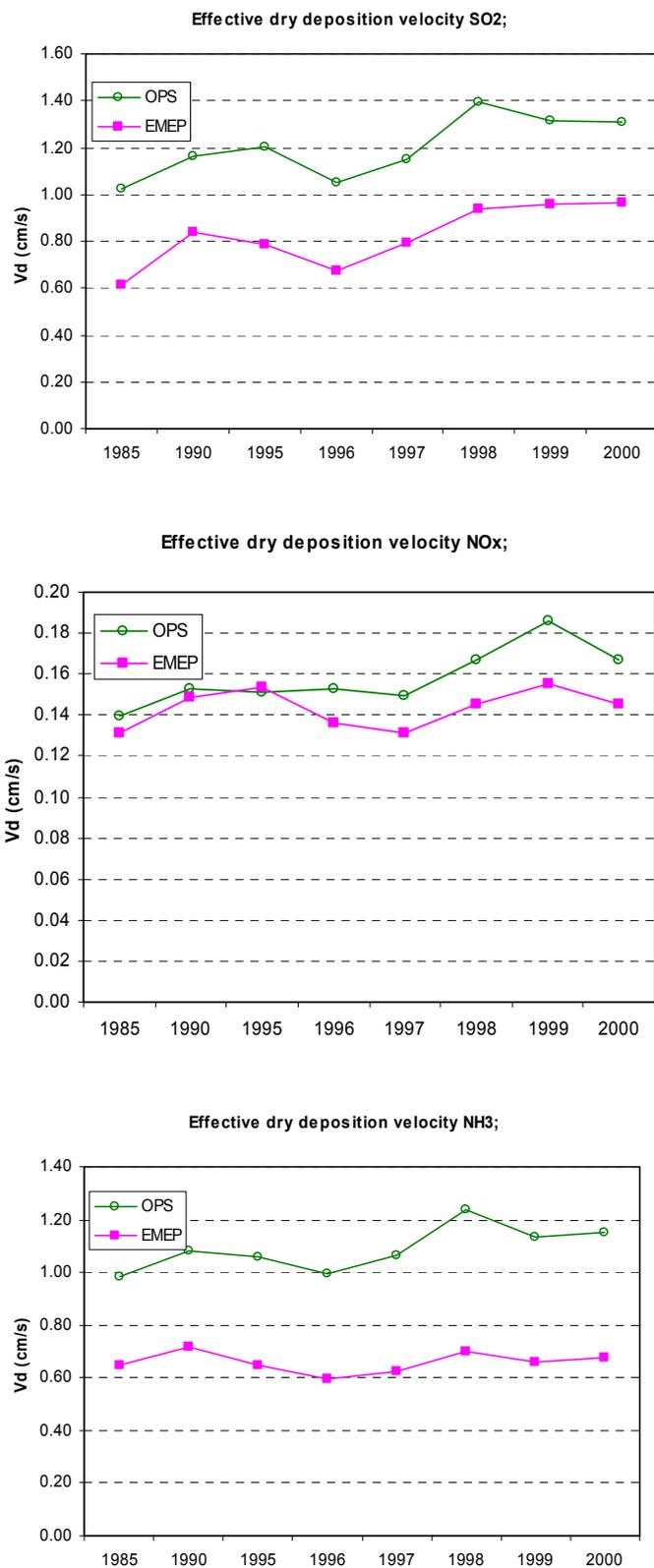
Figure 2.2. Comparison of annual average (for the Netherlands) effective dry deposition velocities for the EMEP and OPS models.

In Figure 2.3 the value of R_{ns} is given as a function of the relative humidity for average conditions in the Netherlands (molar SO_2/NH_3 ratio = 0.09, temperature = $9^{o}C$). The OPS model has a lower R_{ns} under all conditions. For wet surfaces the OPS model uses a value of 5 s/m, while the EMEP value has a lower limit of 17 s/m. Assuming that the sum of R_a and R_b has a value of 50 s/m, V_d has a value of 0.018 m/s for the OPS model and 0.015 for the EMEP model. In case of a relative humidity of 90% these values are 0.018 and 0.077 s/m, respectively. Except for the dependency on humidity, the approaches differ, also in their dependencies on parameters such as temperature and molar SO_2/NH_3 ratios. However, the essential difference between the dry deposition parameterisation in the two models is shown in Figure 2.3; it explains to a large extent why the effective dry deposition velocity in Figure 2.2 (bottom panel) deviates so much between the two models.
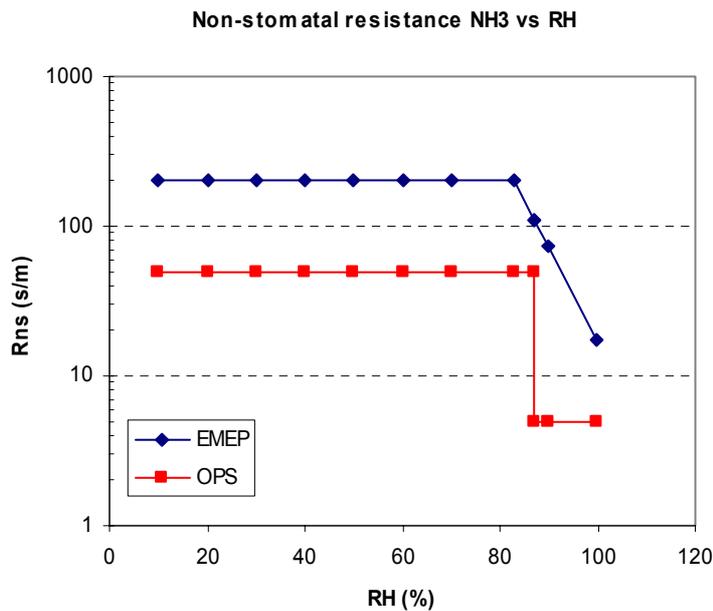
Figure 2.3. Comparison of the non-stomatal resistance formulations for NH₃.
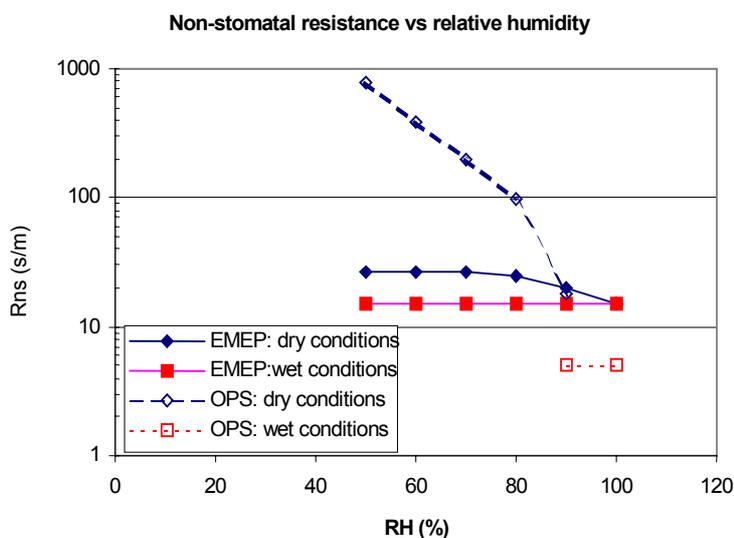
Figure 2.4. Comparison of the non-stomatal resistance formulations for SO₂.

A similar difference between the EMEP and OPS models exists for the non-stomatal resistance of SO_2. Here, the EMEP model recognises two conditions: dry and wet, while in both conditions the resistance varies as a function of the molar SO_2/NH_3 ratio. The 'dry' resistance approaches the wet resistance at 100% relative humidity. The DEPAC parameterisation as applied in the OPS model considers a 'wet' condition at a relative humidity of more than 87%. R_{ns} is independent of the SO_2/NH_3 ratio. In the EMEP model the 'wet' condition exists if there is actual rainfall. The frequency of occurrence of 'wet' is likely to be much higher in the OPS model than in the EMEP model. This leads to lower R_{ns} (on average) and, consequently, higher dry deposition velocities.

Given the discrepancies and considering the impact of high V_d values on SO_2 and NH_3 transport and deposition, it is recommended to re-evaluate R_{ns} formulations (especially for wet conditions) on the basis of original experimental data.

2.2 General comments on dry deposition parameterisations

Dry deposition determines to a large extent the long-range transport capabilities of pollutants and at the same time the atmospheric load of the pollutants to ecosystems. Therefore, source-receptor matrices calculated by long-range transport models are sensitive to the choice of dry deposition parameters (see section 4).

The non-stomatal resistance for ozone is formulated in the EMEP model in a more complex way than for SO_2 or NH_3. The reason mentioned for this is that the ozone case has been evaluated more extensively. The resistance model for ozone includes an additional dry deposition pathway, namely via the canopy through the soil or ground cover. One of the variables is the Leaf Area Index, a function of vegetation type. Basically, there is no difference between dry deposition pathways for the various substances. A uniform approach could make parameterisations more transparent, also in terms of the differences between land use types.

The non-stomatal resistance formulation for NH_3 in the EMEP model contains dependencies on temperature, molar SO_2/NH_3 ratios and humidity. One should expect similar dependencies for SO_2 and other soluble gases because the same processes and the same interaction are concerned. In the EMEP model the formulation for SO_2 is much simpler, while probably much more literature data is available. When it comes to non-linearity issues, the dry deposition dependency between NH_3 and SO_2 may play an important role in future.

The non-stomatal resistance formulation for NH_3 in the EMEP model describes more or less the expected dependencies on parameters such as temperature, humidity and the pollution climate. All these parameters are theoretically defendable and to some extent observed in the laboratory. However, current field experiments are not distinctive enough to positively support a reliable quantification of the necessary parameters.

3.   EMEP versus OPS on a 50 x 50 km grid

The grid size of the EMEP model has decreased from about 150 x 150 km to a current resolution of about 50 x 50 km (the EMEP grid cells are exactly 50 x 50 km but only at 60ºN). The EMEP observational network used to validate the model are located far apart on background sites in Europe. These observations alone might not be sufficient to validate the variability of the EMEP model on the 50 x 50 km grid scale. To test the performance of the EMEP model on the 50 x 50 km grid, we compared its results with output from the OPS model (Van Jaarsveld, 1995) for yearly average concentrations of SO_2, NO_x, and NH_3, as well as dry and wet depositions of oxidised sulphur, oxidised nitrogen and reduced nitrogen for the Netherlands for 1985, 1990 and 1995-2000. The EMEP results are also compared with observations for the Netherlands.

In the Netherlands the OPS model is used for the calculation concentrations and deposition of acidifying compounds. By combining a Gaussian plume model for local transport using a trajectory model for the long-range transport, the model is capable of calculating the effects of very local emission sources (< 100 m), as well as contributions from foreign sources, for example. The model has been validated by comparing SO_2, NO_x and NH_3 concentrations and depositions with measurements from the RIVM's Netherlands air quality monitoring network (LML) (Elzakker, 2001).

To compare the OPS model results (RIVM, 2003) with the EMEP model results, the OPS model results on a 5 x 5 km grid resolution were converted to the EMEP grid format on a 50 x 50 km resolution. The conversion was done as follows: the average of all the OPS grid cells was taken for each EMEP grid cell. Of the cell centres lying in the EMEP grid cell, only cells of which more than 45% of the area of the EMEP grid cell occurred within the OPS area were considered for the comparison; the average and standard deviation in the OPS results was calculated. Background values were added to the dry and wet depositions calculated by the OPS model to account for emissions from outside Europe and the North Sea. The background values for the dry and wet deposition of oxidised sulphur were 38 and 134 mg S/m^2, respectively. For the dry and wet deposition of oxidised nitrogen, these were 18 and 50 mg N/m^2, respectively, and for the dry and wet deposition of reduced nitrogen, 67 and 38 mg N/m^2, respectively.

As an example of the conversion, the concentrations of NH_3 in air are shown in Figure 3.1. The top panel in Figure 3.1 shows the NH_3 concentration on the OPS grid of 5 x 5 km and the bottom left panel shows the OPS concentration on the EMEP grid of 50 x 50 km. For comparison, the bottom right panel of Figure 3.1 shows the NH_3 concentrations of the EMEP model.

In Figure 3.2, the concentrations and depositions calculated with the OPS model are compared with those of the EMEP model per grid cell for the year 2000, being representative of all years between1985-2000 with respect to the correlation and the slope of the least-squares fit of the correlation's. In general, there is a large dispersion in the converted OPS results for each EMEP grid cell, especially at high concentrations (Figure 3.2). For SO_2 and NH_3, the EMEP and OPS concentrations agree quite well, the EMEP concentrations are slightly higher for most grid cells. For NO_x, the OPS model calculates much higher concentrations for each cell than the EMEP model, almost a factor of two. This underestimation of the NO_x concentration in the EMEP model might be related to the fact that Eulerian (grid) models often have difficulties with the large spatial gradients in NO_x close to emission sources. After being emitted, NO_x is distributed over the whole grid cell, resulting in a larger reduction in concentration than in reality.

The dry deposition (Figure 3.2) calculated by the EMEP model is much lower than that calculated by the OPS model for oxidised and reduced nitrogen. For most grid cells, the dry deposition of the EMEP model is, for oxidised sulphur, lower than that of the OPS model, but for low OPS values, both agree quite well. The wet deposition calculated by the EMEP model is always much higher than in the OPS model for all three components.

In Tables 3.1 to 3.4 and Figures 3.3 to 3.6 the averaged concentrations and depositions of the EMEP model are compared with those of the OPS model and with measurements. For comparing the averaged concentrations and depositions the data are averaged over the area of the Netherlands. The average values are calculated using a weighted mean of the fraction of the EMEP cell covering the Netherlands. The measurements are taken from the RIVM's Netherlands air quality monitoring network (LML) (Elzakker, 2001).
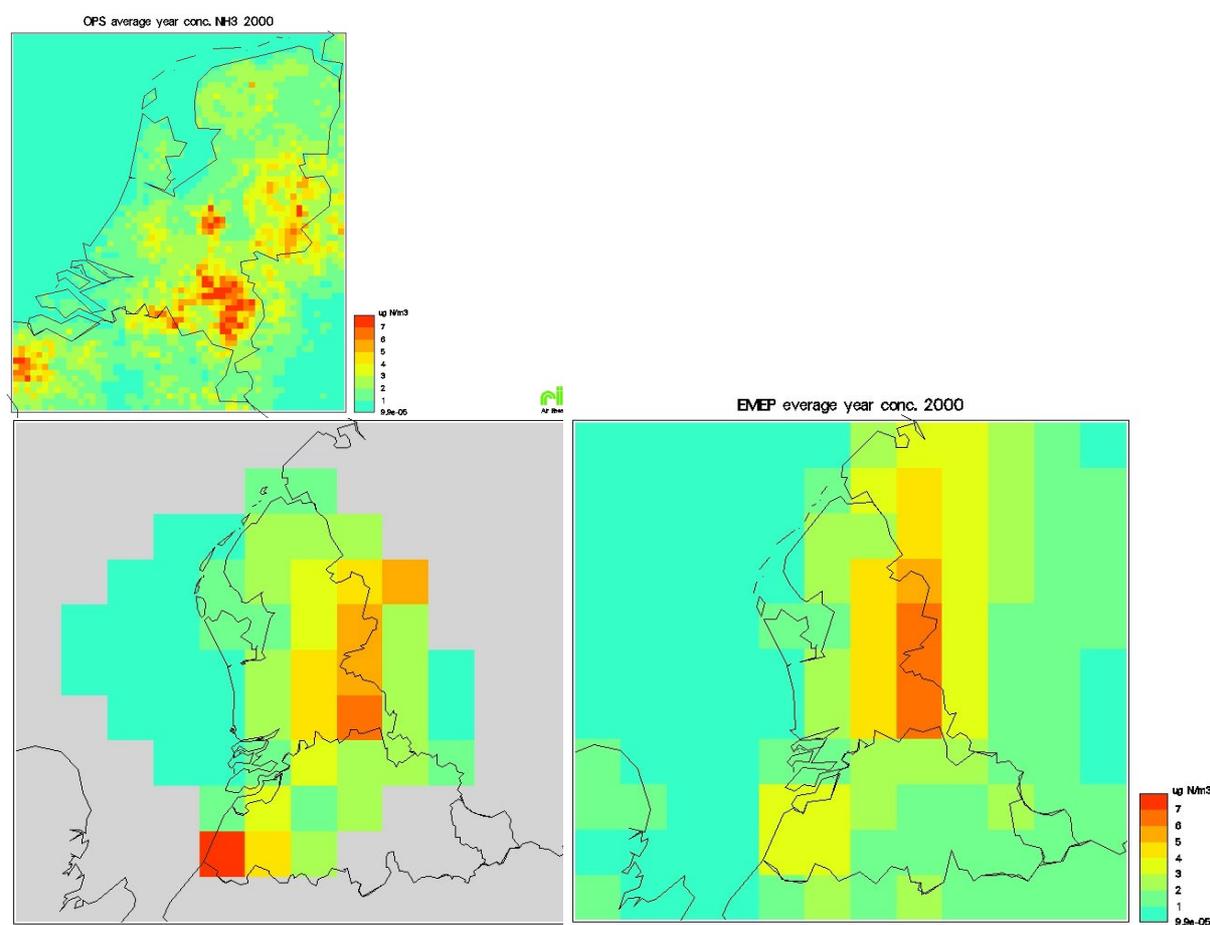
Figure 3.1. Air concentration of NH₃ (in μg m⁻³) in the Netherlands from the OPS model (top panel), from the OPS model converted to the EMEP grid (bottom left panel), and from the EMEP model (bottom right panel).
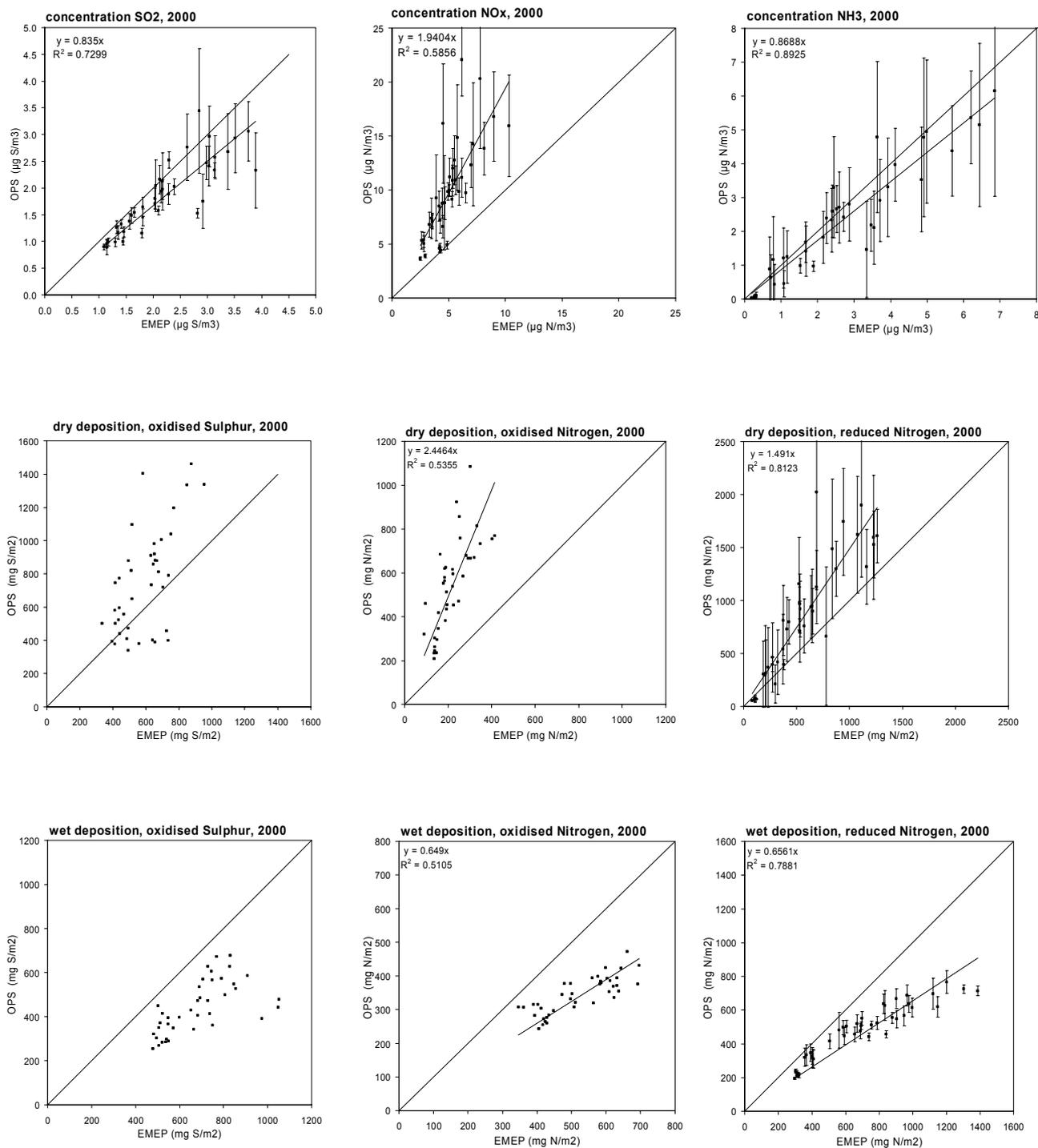
Figure 3.2. Correlation between the EMEP and OPS results for the concentrations in air of SO₂, NOₓ and NH₃ and the dry and wet deposition of oxidised sulphur (SOₓ) along with oxidised (NOᵧ) and reduced (NHₓ) nitrogen in the Netherlands for 2000. The OPS results were converted to the EMEP grid. The vertical bars represent the variability (standard deviation) in the OPS grid cells.

Table 3.1. Concentration[1] of SO$_2$ (µg S m^{-3}), NO$_x$ and NH$_3$ (µg N m^{-3}) in the Netherlands from the OPS model, the Eulerian EMEP model and LML observations[2].

	SO$_2$			NO$_x$			NH$_3$		
year	EMEP	OPS[3]	LML	EMEP	OPS[3]	LML	EMEP	OPS[3]	LML
1985	8.42	6.53 (0.74)	9.88	8.10	17.8 (3.6)	16.2	4.69	3.79 (1.58)	-
1990	4.94	4.00 (0.50)	4.51	6.93	14.2 (2.8)	11.9	4.26	4.27 (1.78)	-
1995	3.42	2.51 (0.32)	2.50	6.10	12.6 (2.5)	10.7	3.92	3.93 (1.66)	6.2
1996	3.57	2.85 (0.31)	3.21	6.46	13.0 (2.5)	11.7	3.39	3.49 (1.35)	6.5
1997	2.72	2.23 (0.28)	2.15	6.26	12.5 (2.4)	11.3	4.19	3.66 (1.48)	6.6
1998	2.14	1.81 (0.26)	1.50	5.49	10.7 (1.9)	9.3	3.45	3.06 (1.23)	4.6
1999	1.81	1.67 (0.24)	1.57	5.15	10.0 (2.0)	8.4	3.51	3.55 (1.46)	5.2
2000	1.85	1.60 (0.22)	1.54	5.43	10.2 (1.9)	8.2	3.38	3.07 (1.26)	4.8

Table 3.2. Dry deposition[1] of oxidised sulphur (mg S m^{-2}) and oxidised and reduced nitrogen (mg N m^{-2}) in the Netherlands from the OPS model and the Eulerian EMEP model.

	oxidised Sulphur, SO$_x$			oxidised Nitrogen, NO$_y$			reduced Nitrogen, NH$_x$		
year	EMEP	OPS[3]	LML	EMEP	OPS[3]	LML	EMEP	OPS[3]	LML
1985	1635	2113 (799)	-	334	784 (143)	-	955	1177 (365)	-
1990	1313	1465 (618)	-	325	684 (120)	-	961	1459 (451)	-
1995	852	953 (373)	-	295	599 (108)	-	799	1315 (415)	-
1996	760	947 (349)	-	278	627 (113)	-	634	1098 (333)	-
1997	679	808 (303)	-	259	589 (105)	-	824	1228 (381)	-
1998	633	796 (317)	-	251	564 (100)	-	760	1195 (377)	-
1999	546	693 (272)	-	252	586 (106)	-	732	1269 (405)	-
2000	563	661 (253)	-	248	536 (95)	-	723	1113 (357)	-

Table 3.3. Wet deposition[1] of oxidised sulphur (mg S m^{-2}) and oxidised and reduced nitrogen (mg N m^{-2}) in the Netherlands from the OPS model, the Eulerian EMEP model and LML observations[1].

	oxidised Sulphur, SO$_x$			oxidised Nitrogen, NO$_y$			reduced Nitrogen, NH$_x$		
year	EMEP	OPS[3]	LML	EMEP	OPS[3]	LML	EMEP	OPS[3]	LML
1985	2095	1159 (85)	1069	754	517 (16)	498	1048	736 (72)	990
1990	1275	887 (63)	982	537	427 (12)	428	717	627 (70)	934
1995	863	660 (43)	701	473	393 (11)	412	643	482 (54)	846
1996	738	533 (33)	579	466	340 (7)	364	615	411 (39)	783
1997	643	535 (38)	608	473	334 (7)	363	688	466 (46)	846
1998	886	683 (55)	816	640	424 (11)	511	907	590 (62)	1021
1999	664	577 (44)	643	501	431 (13)	449	737	525 (52)	864
2000	680	573 (43)	682	533	386 (11)	518	789	515 (50)	939

Table 3.4. Total (dry + wet) deposition[1] of oxidised sulphur (mg S m^{-2}) and oxidised and reduced nitrogen (mg N m^{-2}) in the Netherlands from the OPS model and the Eulerian EMEP model.

	oxidised Sulphur, SO$_x$			oxidised Nitrogen, NO$_y$			reduced Nitrogen, NH$_x$		
year	EMEP	OPS	LML	EMEP	OPS	LML	EMEP	OPS	LML
1985	3730	3273	-	1088	1302	-	2003	1913	-
1990	2588	2353	-	862	1112	-	1678	2086	-
1995	1715	1614	-	768	993	-	1442	1797	-
1996	1498	1481	-	744	968	-	1249	1509	-
1997	1322	1344	-	732	924	-	1512	1694	-
1998	1519	1480	-	891	989	-	1667	1785	-
1999	1210	1271	-	753	1018	-	1469	1794	-
2000	1243	1235	-	781	923	-	1512	1628	-

1) Values are averaged over the area of the Netherlands using the EMEP grid.
2) Observations from the LML network (Elzakker, 2001) represent the average for the area of the Netherlands.
3) The average variability (standard deviation of the average) in the OPS grid cells mapped onto the EMEP grid is given in parentheses.
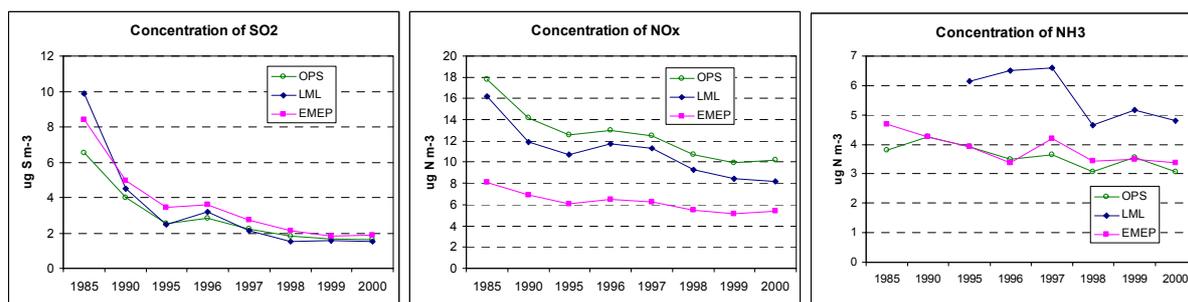
Figure 3.3. Comparison of the concentrations in air of SO₂, NOₓ and NH₃ in the Netherlands for the EMEP and OPS models and the LML measurements.
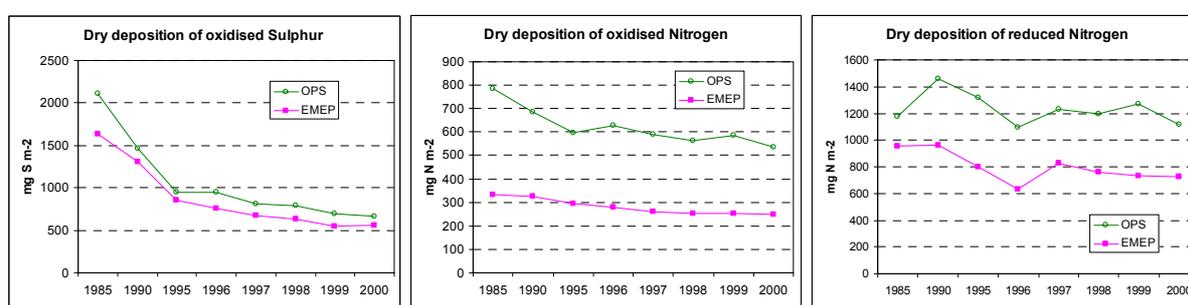
Figure 3.4. Comparison of the dry deposition of oxidised sulphur and oxidised and reduced nitrogen in the Netherlands for the EMEP and OPS models.
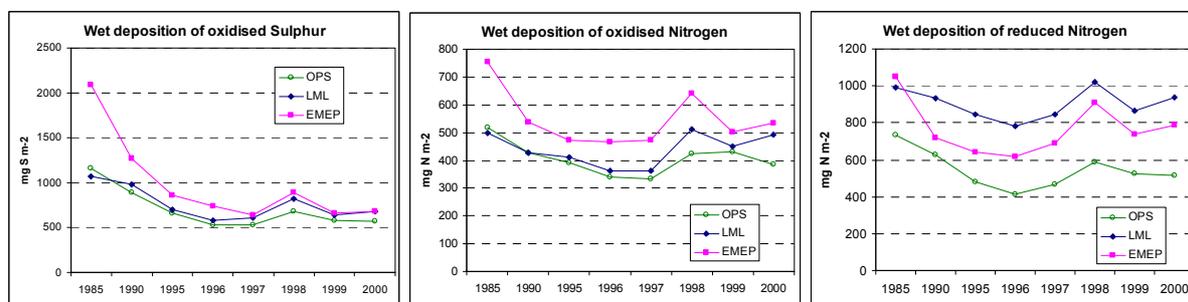
Figure 3.5. Comparison of the wet deposition of oxidised sulphur and oxidised and reduced nitrogen in the Netherlands for the EMEP and OPS models and the LML measurements.
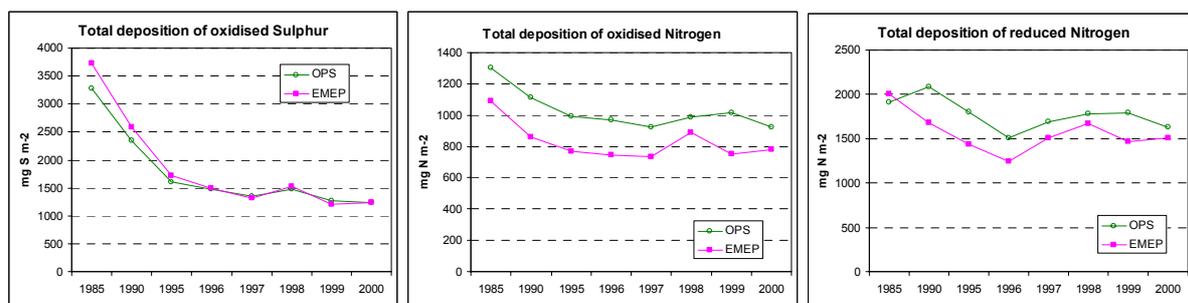
Figure 3.6. Comparison of the total (dry + wet) deposition of oxidised sulphur and oxidised and reduced nitrogen in the Netherlands for the EMEP and OPS models.

Comparing the yearly averaged SO_2 concentration in the Netherlands (Table 3.1 and Figure 3.3, we found that concentrations calculated with the EMEP model for the later years were close to the OPS results and the measurements. The EMEP values are always slightly higher than the OPS results and the measurements, except for 1985 when the EMEP model yielded values about 15% below the measurements. The NO_x concentrations calculated with the EMEP model are about 50% lower than the OPS concentrations and about 40% lower than the measured concentrations for all the years. This underestimation of the NO_x concentrations by the EMEP model, compared with the measurements, is in agreement with the EMEP report (Simpson *et al*., 2003). They find a somewhat smaller underestimation by considering two stations (Kollumerwaard and Vredepeel) with for Dutch standards low NO_x concentrations. For the NH_3 concentrations, the EMEP results are close to the OPS results, but both are 30 to 40% lower than the measured concentrations. This occurrence, in combination with the fact that both models underestimate the wet deposition of NH_x, confirms the existence of the so-called 'ammonia gap' (Van Jaarsveld and Van Pul, 2002).

The dry deposition calculated with the EMEP model is lower than that calculated with the OPS model for all compounds, while the wet deposition is always higher. Consequently, the total (dry + wet) deposition of both models are similar, especially for oxidised sulphur. The dry deposition of the EMEP model is lower than the OPS model, by an average of about 15%, 55%, and 35%, for oxidised sulphur, oxidised nitrogen, and reduced nitrogen, respectively. There are no measurements to compare these dry depositions with. The dry deposition is determined by the concentration in the air and the dry deposition velocity (see section 4.1). Using a larger dry deposition, velocity will almost proportionally increase the dry deposition, but result in a (small) decrease in the atmospheric concentration. Due to conservation of mass, the wet deposition decreases if the dry deposition increases, resulting in only a small change in the total deposition.

The wet deposition calculated with the EMEP model is higher for all compounds (about 20-50%) than that calculated with the OPS model. The differences between the EMEP and OPS models for the wet deposition of oxidised sulphur decreased from 81% in 1985 to 19% in 2000. Especially for the later years, there is a good agreement with the measurements. The wet deposition of oxidised and reduced nitrogen of the EMEP model is about 25% higher and about 20% lower, respectively, than the measurements.

The standard deviation around the average values in the OPS grid cells is shown in Figure 3.2 and Tables 3.1 to 3.3 to express the fact that the concentrations and depositions of the OPS grid cells on a 5 x 5 km grid scale exhibit considerable variation. Closer investigation of this variability shows that it is about a constant percentage of the average value and that there is no trend over the years in this variability. For example, the standard deviation of the OPS grid cells for the SO_2 concentration in air is on average about 12% for all years.

4. Source-receptor relationships for the Netherlands

The OPS model (Van Jaarsveld, 1995) is used at RIVM-MNP to calculate source-receptor relationships for NO_x, SO_x and NH_x deposition. A full evaluation of the source-receptor relationship for Europe requires a large number of scenario calculations with a European model due to the non-linearity of the processes involved. This is outside the scope of this analysis and of the current version of the OPS model. This chapter compares the contribution of countries to the deposition in the Netherlands, as calculated by the EMEP and OPS models. For this purpose the year 2000 is selected. The EMEP data is taken from the EMEP Status Report 1/2003 Part III (Tarrasón *et al.*, 2003) and the OPS data from the calculations carried out in the framework of the Netherlands Environmental Balance 2003 (RIVM, 2003).

The OPS data are calculated on the basis of the same national emission totals as used in the EMEP model; however, the spatial distribution was not the same nor were the physical properties of the emission sources (release height, heat content, spatial resolution). The spatial resolution for emissions in the Netherlands, Belgium and Germany was 5 x 5 km, while large point source emissions were treated as point sources conforming with their exact locations. Separate 5 x 5 km distributions were used for the remaining (non-point source) emissions, segregated into specific categories (industrial, mobile sources, inland shipping, agricultural sources etc.).

The comparison is based on the sum of the deposition on the Netherlands territory. Because the area considered in the two models is not the same surface area (OPS: 34958 km^2, EMEP: 42485 km^2), the OPS data will be systematically lower than the EMEP data. However, a simple scaling on the basis of the respective surface areas would lead to an overestimation, because the EMEP area includes more of the surrounding waters where deposition is lower than on the land area. In order to be compatible with the comparison in section 3, we have not applied a correction factor. The comparison on the total deposition may therefore differ from what is given in section 3. Results are given in Table 4.2 and Figure 4.1

One difference in approach between the EMEP and the OPS models is that the Eulerian EMEP model applies a much larger Boundary Initial Conditions (BIC) at the model domain boundaries than the OPS model. One can consider them as background concentrations accounting for trans-Atlantic transport, for example. The initial concentrations represent, in fact, a source term and signify a certain contribution to the Netherlands. A similar problem applies to the OPS model. Since only anthropogenic emissions in Europe are considered and no shipping emissions are taken into account (except North Sea emissions), background contributions are added to the model results. These depositions are based on estimates made by Locht and Van Aalst (1988). Table 4.1 provides the values for the different compounds.

Table 4.1. Background depositions applied to the Netherlands.

	Unit	dry	wet	total
NO_y	mg N m^{-2} a^{-1}	18	50	69
NH_x	mg N m^{-2} a^{-1}	67	38	105
SO_x	mg S m^{-2} a^{-1}	38	134	173

Table 4.2. Deposition contributions to the Netherlands[1] according to the EMEP and the OPS models for the year 2000.

	SO_x (100 tonne S)		NO_y (100 tonne N)		NH_x (100 tonne N)	
	EMEP	**OPS**	**EMEP**	**OPS**	**EMEP**	**OPS**
NL	84	108	28	121	304	460
BE	76	70	15	26	81	47
DE	41	41	22	41	57	45
FR	45	34	40	29	52	17
GB	72	63	61	44	15	11
other Eur. countries	36	27	28	21	18	5
NOS[2]	63	62	32	34	-4	0
BAS[2]	1		1			
MED[2]	1		1			
ATL[2]	5		9			
BIC[2]	13	61#	103	24#		37#
Total	437	463	340	340	523	622

1) The grid and total area used in the EMEP model differs here from the area used in the OPS model, which gives a difference in the absolute values between the models. Furthermore, figures in this table can not be directly compared with those in section 3 (Figure 3.6 and Table 3.4).
2) NOS = North Sea; BAS = Baltic Sea; MED = Mediterranean; ATL = Atlantic; BIC = Boundary and Initial Conditions
#) Background value, accounting for non-European and natural source contributions (see section 3).

The following observations can be made with respect to the source-receptor matrices (Table 4.2 and Figure 4.1):

- The source-receptor matrices for SO_x calculated by the two models are in good agreement.

- The agreement for reduced nitrogen is reasonable. The EMEP model calculates larger contributions from Belgium and Germany and a lower contribution from the Netherlands.

- The relative contribution from the Netherlands to the Netherlands is always higher in the OPS model than in the EMEP model: for SO_x EMEP is 20% and OPS: 24%), for NH_x, EMEP is 58% and OPS 73% and for NO_y EMEP is 8% and OPS 36%). This is in line with the findings in chapter 3 that the OPS model calculates higher concentrations (NO_x) and/or uses higher dry deposition velocities (SO_2 and NH_3; see also chapter 2, Figure 2.1).

- The largest discrepancies are found for oxidised nitrogen. Here, the local deposition contribution of Dutch emissions to the Netherlands is a factor of 4 higher than in the EMEP model. The Dutch emissions contribute only 8% to the emissions in the Netherlands. This was 30-35% in the pervious version of the Eulerian model and 15-20% in the Lagrangian model (EMEP, 2001). The NO_y contributions from Belgium and Germany are now also much lower in the EMEP model than in the OPS model. A comparison of model results with observations in the Netherlands (chapter 3) illustrates that the EMEP model strongly underestimates NO_x concentrations and dry deposition, while the wet deposition is overestimated. This is not related to the combination of low sources and a low vertical resolution of the model, as can be seen in the case of NH_3 where local concentrations are much less underestimated.

- Another striking difference is the influence of BIC for oxidised nitrogen. The EMEP model suggests that almost 30% of the deposition is due to sources outside Europe. In the previous version of the EMEP Eulerian model, this BIC term contributed only 3-5% to the deposition of NO_y in the Netherlands. Apparently, the EMEP model has difficulties modelling oxidised nitrogen.

- The EMEP model reports no contributions of BIC for NH_x. It is unclear if these are either not taken into account or are not significant.

At the TFMM workshop in Oslo in November 2003 the data of Table 4.2 was discussed with EMEP. They explained that the SRMs calculated by the EMEP UNIFIED model (EMEP Status Report 1/2003 Part III, Tarrasón *et al*., 2003) are not the final results. They explained that the large BIC term for NO_y is not a true boundary condition, but comes from the way the SRMs for NO_y are calculated. They are still working on the SRM calculations and expect to have SRMs ready for integrated assessment studies by May 2004.
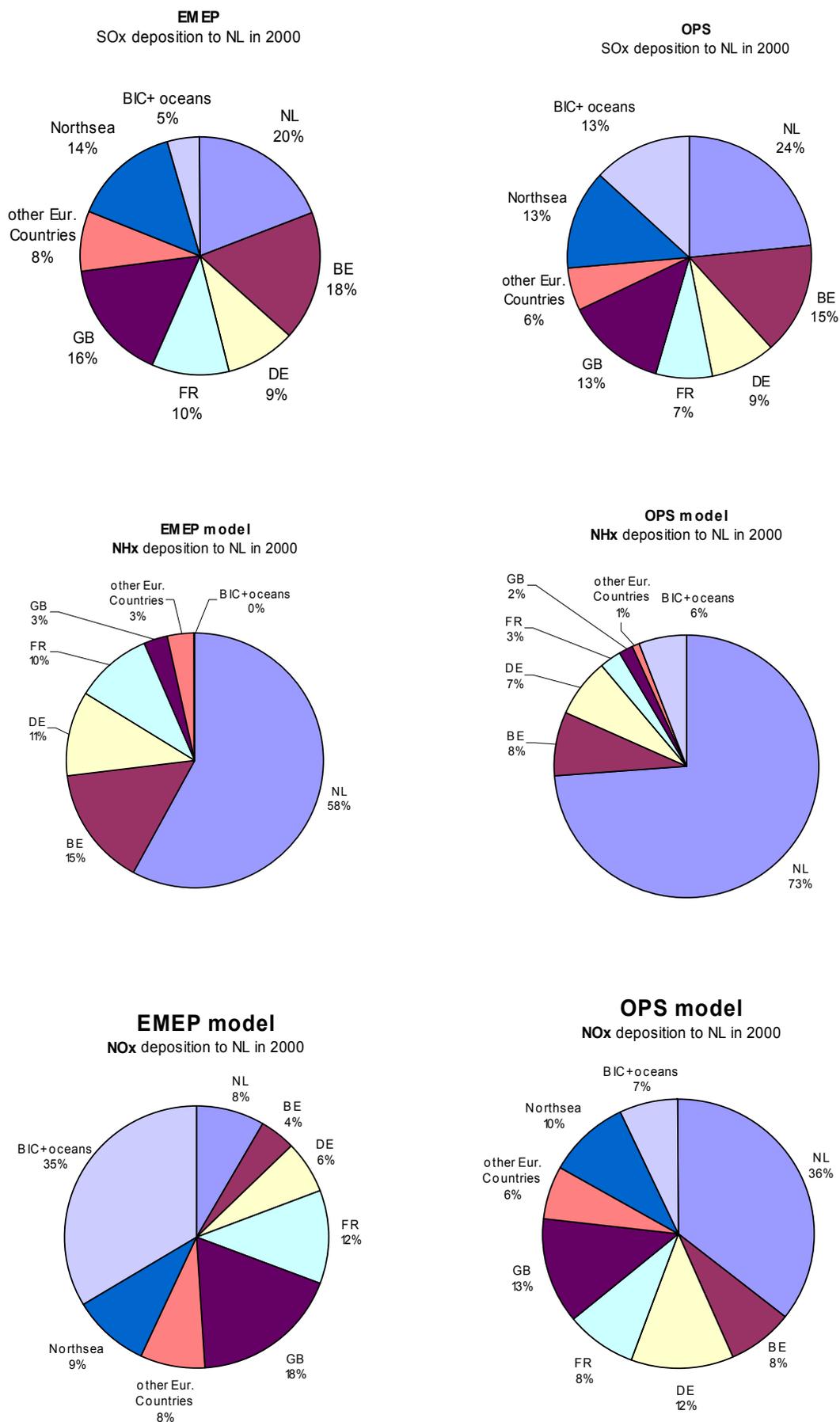
Figure 4.1. Deposition contributions to the Netherlands according to the EMEP and OPS models for the year 2000.

5. Distribution of emissions

In Task I of the evaluation EMEP compared the model results with measurements for 1980-2000. This comparison depends strongly on the emissions and their geographical distribution as used in the EMEP model. Countries were requested to report their emissions and the geographical distribution of the emissions to EMEP. We have compared the geographical distributions of the EMEP's SO_x and NO_x emissions on the 50 x 50 km grid for selected countries, i.e., Germany, The Netherlands and Poland and selected years, 1980 and from 1985-2000. In Germany and Poland significant changes in the geographical distribution of the SO_x and NO_x emissions from 1980 to 2000 are be expected as a result of changes in the economies of these countries. Investigated were the low and high emission categories and the emissions per sector.

Tables 5.1 and 5.2 show the SO_x and NO_x emissions for the low and high emission categories, as obtained from the EMEP website (Webdab, 2002). The ratios between the emissions in both categories are shown as the percentage emitted per grid cell (compared with the total emission in the country). It can be seen that:

1. the ratio between the emissions in the high and low emission categories is, per grid cell, the same for all the years in the period 1980-2000 (i.e., 1980, 1985-2000). This holds for all grid cells in the three selected countries and for both the SO_x and NO_x emissions.

2. the geographical distribution of the SO_x and NO_x emissions (low and high emission categories) in the selected countries is identical for all of 1980 and 1985-2000, i.e. the same fraction of the total emissions in a country is emitted in the grid cells for all years.

3. the ratio between emissions in the high and low emission categories for Poland is the same for all cells and all years.

The EMEP emissions are also subdivided in 11 sectors, e.g. combustion, transport, etc. The percentage of the emissions in the grid cells, compared with the total emission in the country, is analysed for Germany, The Netherlands and Poland for the years 1980 and 1985-2000. The data for Germany are shown in Table 5.3 for the three sectors with the largest emissions and for the years 1980 and 2000. From this table it can be seen that:

4. the geographical distribution of the SO_x and NO_x emissions of all sectors in the selected countries does not change through the years 1980-2000, i.e. the same fraction of the total emissions of a sector in a country is emitted in the grid cells for all years.

The absence of changes in geographical distribution in the EMEP emissions from 1980 to 2000 will likely affect the comparison between modelled and measured concentrations. This will hold especially for Eastern Europe, where large changes in emissions and their geographical distributions are expected due to the changes in their economies since the 1980s. The effects for Western Europe will probably be much smaller.

The emissions discussed here are obtained from the EMEP website (Webdab, 2002). By the end of October 2003 EMEP released the new version of the emission database, Webdab 2003. An analysis of the new emissions showed that, although some numbers have changed, the conclusions about the constant geographical distributions, are still valid. The emissions show changes in geographical distributions only for the years 2000 and 2001.

In a discussion, after this report was placed on the EMEP website, EMEP explained that the low and high emissions of Webdab 2002 have not been used for the Unified model. They also explained that, although countries are requested to report geographical distributions of their emissions, many don't send this information to EMEP and EMEP uses what is available.

Table 5.1. Geographical distribution of SO_x emissions on the EMEP 50 x 50 km grid for Germany, The Netherlands and Poland for 1980 and 2000, showing the emissions (in Mg SO_2) in the low and high emission categories for the grid cells with the largest emissions (in 1980), and the ratio between the high and low emission categories and the percentage of the emissions in the grid cells compared with the total emission in the country[1].

Country	Cell		Low emissions		High emissions		Ratio high/low		Percentage total: low		Percentage total: high	
	i	j	1980	2000	1980	2000	1980	2000	1980	2000	1980	2000
Germany	60	45	234879	25976	352210	38952	1.4995	1.4995	5.7292	5.7292	10.3157	10.3157
Germany	61	45	136589	15106	92078	10183	0.6741	0.6741	3.3317	3.3317	2.6968	2.6968
Germany	60	46	124948	13818	154475	17084	1.2363	1.2363	3.0477	3.0477	4.5244	4.5244
Germany	61	46	112395	12430	73216	8097	0.6514	0.6514	2.7415	2.7415	2.1444	2.1444
Germany	60	52	96708	10695	72136	7978	0.7459	0.7459	2.3589	2.3589	2.1128	2.1128
Germany	65	43	83028	9182	49864	5515	0.6006	0.6006	2.0252	2.0252	1.4604	1.4604
Germany	61	44	78670	8700	71918	7954	0.9142	0.9142	1.9189	1.9189	2.1064	2.1064
Germany	67	43	76088	8415	61372	6787	0.8066	0.8066	1.8559	1.8559	1.7975	1.7975
Germany	64	44	71721	7932	53222	5886	0.7421	0.7421	1.7494	1.7494	1.5588	1.5588
Germany	71	44	68892	7619	48828	5400	0.7088	0.7088	1.6804	1.6804	1.4301	1.4301
Country total[2]			4099704	453401	3414296	377599						
Netherlands	57	44	92701	17254	145556	27091	1.5702	1.5702	35.2373	35.2373	64.1433	64.1433
Netherlands	57	43	31317	5829	29165	5428	0.9313	0.9313	11.9042	11.9042	12.8525	12.8525
Netherlands	57	42	18745	3489	695	129	0.0371	0.0371	7.1254	7.1254	0.3062	0.3062
Netherlands	56	45	15465	2878	4896	911	0.3166	0.3166	5.8784	5.8784	2.1577	2.1577
Netherlands	58	44	13703	2550	14453	2690	1.0548	1.0548	5.2087	5.2087	6.3693	6.3693
Netherlands	57	45	13334	2482	7201	1340	0.5401	0.5401	5.0685	5.0685	3.1735	3.1735
Netherlands	58	45	10947	2038	934	174	0.0853	0.0853	4.1612	4.1612	0.4115	0.4115
Netherlands	56	44	8695	1618	14474	2694	1.6646	1.6646	3.3052	3.3052	6.3785	6.3785
Netherlands	59	45	6100	1135	586	109	0.0960	0.0960	2.3187	2.3187	0.2580	0.2580
Netherlands	58	49	5930	1104	1215	226	0.2048	0.2048	2.2541	2.2541	0.5352	0.5352
Country total[2]			263077	48965	226923	42236						
Poland	75	56	250368	92270	212482	78308	0.8487	0.8487	11.2890	11.2890	11.2890	11.2890
Poland	73	58	242118	89229	205480	75727	0.8487	0.8487	10.9170	10.9170	10.9170	10.9170
Poland	70	52	120212	44303	102021	37599	0.8487	0.8487	5.4203	5.4203	5.4203	5.4203
Poland	75	55	116361	42883	98753	36394	0.8487	0.8487	5.2467	5.2467	5.2467	5.2467
Poland	70	58	91747	33812	77864	28696	0.8487	0.8487	4.1369	4.1369	4.1369	4.1369
Poland	65	56	72741	26808	61734	22751	0.8487	0.8487	3.2799	3.2799	3.2799	3.2799
Poland	73	61	70808	26095	60094	22147	0.8487	0.8487	3.1927	3.1927	3.1927	3.1927
Poland	76	59	70365	25932	59717	22008	0.8487	0.8487	3.1727	3.1727	3.1727	3.1727
Poland	76	57	62235	22936	52818	19465	0.8487	0.8487	2.8062	2.8062	2.8062	2.8062
Poland	74	61	57052	21026	48419	17844	0.8487	0.8487	2.5725	2.5725	2.5725	2.5725
Country total[2]			2217799	817340	1882201	693660						

1) The data of only a few grid cells per country and two years are shown, but the same results can be seen for all grid cells and all the years in 1980 and 1985-2000.
2) The country total is calculated over all grid cells in a country in general, not only the ones shown here.

Table 5.2. Geographical distribution of NO_x emissions on the EMEP 50 x 50 km grid for Germany, The Netherlands, and Poland for 1980 and 2000. Shown are the emissions (in Mg NO_2) in the low and high emission categories for the grid cells with the largest emissions (in 1980), as well as the ratio between the high and low emission categories and the percentage of the emissions in the grid cells compared with the total emission in the country[1].

Country	Cell		Low emissions		High emissions		Ratio high/low		Percentage total: low		Percentage total: high	
	i	j	1980	2000	1980	2000	1980	2000	1980	2000	1980	2000
Germany	60	45	85061	41765	36342	17844	0.4272	0.4272	2.9679	2.9679	7.7657	7.7657
Germany	61	45	65450	32136	12979	6373	0.1983	0.1983	2.2837	2.2837	2.7734	2.7734
Germany	61	46	55497	27249	10562	5186	0.1903	0.1903	1.9364	1.9364	2.2570	2.2570
Germany	64	44	49668	24387	7246	3558	0.1459	0.1459	1.7330	1.7330	1.5483	1.5483
Germany	67	43	48680	23902	8949	4394	0.1838	0.1838	1.6985	1.6985	1.9123	1.9123
Germany	60	46	47831	23485	19175	9415	0.4009	0.4009	1.6689	1.6689	4.0975	4.0975
Germany	61	44	44506	21852	9987	4903	0.2244	0.2244	1.5529	1.5529	2.1340	2.1340
Germany	71	44	44408	21805	7068	3470	0.1592	0.1592	1.5495	1.5495	1.5103	1.5103
Germany	60	52	43159	21191	9891	4856	0.2292	0.2292	1.5059	1.5059	2.1135	2.1135
Germany	64	45	42958	21093	5840	2868	0.1360	0.1360	1.4989	1.4989	1.2480	1.2480
Country total[2]			2866019	1407221	467981	229779						
Netherlands	57	44	91297	65925	18364	13261	0.2011	0.2011	17.9905	17.9905	24.3146	24.3146
Netherlands	57	45	61268	44241	7660	5531	0.1250	0.1250	12.0731	12.0731	10.1417	10.1417
Netherlands	58	45	48299	34877	1973	1425	0.0409	0.0409	9.5176	9.5176	2.6128	2.6128
Netherlands	58	44	39378	28434	7100	5127	0.1803	0.1803	7.7596	7.7596	9.4005	9.4005
Netherlands	59	45	23816	17197	1240	896	0.0521	0.0521	4.6930	4.6930	1.6422	1.6422
Netherlands	57	43	23573	17022	5675	4098	0.2407	0.2407	4.6451	4.6451	7.5139	7.5139
Netherlands	59	44	21094	15232	1442	1041	0.0683	0.0683	4.1568	4.1568	1.9087	1.9087
Netherlands	58	46	20664	14921	908	656	0.0439	0.0439	4.0719	4.0719	1.2022	1.2022
Netherlands	57	42	16761	12103	2000	1444	0.1193	0.1193	3.3029	3.3029	2.6479	2.6479
Netherlands	59	46	15229	10997	876	633	0.0575	0.0575	3.0010	3.0010	1.1600	1.1600
Country total[2]			507473	366442	75527	54538						
Poland	75	56	77320	52721	27588	18811	0.3568	0.3568	8.5360	8.5360	8.5360	8.5360
Poland	73	58	55330	37727	19742	13461	0.3568	0.3568	6.1083	6.1083	6.1083	6.1083
Poland	75	55	51805	35324	18484	12603	0.3568	0.3568	5.7192	5.7192	5.7192	5.7192
Poland	76	57	41156	28062	14684	10013	0.3568	0.3568	4.5436	4.5436	4.5436	4.5436
Poland	73	61	38807	26461	13846	9441	0.3568	0.3568	4.2843	4.2843	4.2843	4.2843
Poland	74	55	23325	15904	8322	5675	0.3568	0.3568	2.5751	2.5751	2.5751	2.5751
Poland	65	56	22067	15046	7873	5368	0.3568	0.3568	2.4361	2.4361	2.4361	2.4361
Poland	70	58	19653	13401	7012	4781	0.3568	0.3568	2.1697	2.1697	2.1697	2.1697
Poland	74	61	18811	12827	6712	4577	0.3568	0.3568	2.0768	2.0768	2.0768	2.0768
Poland	72	59	15300	10432	5459	3722	0.3568	0.3568	1.6891	1.6891	1.6891	1.6891
Country total[2]			905808	617630	323192	220370						

1) The data of only a few grid cells per country and two years are shown, but the same results can be seen for all grid cells and all of 1980 and 1985-2000.
2) The country total is calculated over all grid cells in a country in general, not only the ones shown here.

Table 5.3. Geographical distribution of SO_x emissions on the EMEP 50 x 50 km grid for the three largest emissions sectors for Germany for 1980 and 2000. Shown are the emissions (in Mg SO_2) for the grid cells with the largest emissions (in 1980, as well as the percentage of the emissions in the grid cells compared with the total emission in the country[1].

Cell		Emissions in 1980			Emissions in 2000			Percentage of country total					
i	j	1 Energy industries	2 Non-industrial	3 Manu-facturing	1 Energy industries	2 Non-industrial	3 Manu-facturing	1 Energy industries		2 Non-industrial		3 Manufacturing	
								1980	2000	1980	2000	1980	2000
60	45	454430	28771	56038	50257	3182	6197	10.8599	10.8599	3.0405	3.0405	4.0402	4.0402
60	46	197472	18198	30717	21839	2013	3397	4.7192	4.7192	1.9231	1.9231	2.2146	2.2146
61	45	108904	30258	51486	12044	3346	5694	2.6026	2.6026	3.1976	3.1976	3.7120	3.7120
61	46	85977	21067	43152	9509	2330	4772	2.0547	2.0547	2.2263	2.2263	3.1112	3.1112
60	52	90066	13039	22379	9961	1442	2475	2.1524	2.1524	1.3780	1.3780	1.6135	1.6135
61	44	90470	12825	19261	10005	1418	2130	2.1620	2.1620	1.3553	1.3553	1.3887	1.3887
63	41	123606	2115	11257	13670	234	1245	2.9539	2.9539	0.2235	0.2235	0.8116	0.8116
67	43	72722	14696	35400	8043	1625	3915	1.7379	1.7379	1.5531	1.5531	2.5523	2.5523
65	43	59269	14064	25680	6555	1555	2840	1.4164	1.4164	1.4862	1.4862	1.8515	1.8515
65	53	88118	2545	24766	9745	281	2739	2.1058	2.1058	0.2689	0.2689	1.7856	1.7856
Total[2]		4184477	946269	1386990	462776	104651	153392						

1) The data of only a few grid cells for some sectors and two years are shown. However, the same results can be seen for all grid cells, for Germany, The Netherlands and Poland, for all 11 sectors and for all of 1980, and 1985-2000, as well as for the NO_x emissions.

2) The country total is calculated over all grid cells in a country in general, not only the ones shown here.

6. Discussion

The EMEP Unified model gives a fair picture of the Netherlands for most acidifying compounds. The source-receptor matrices for oxidised sulphur calculated by the EMEP and OPS models are in good agreement, while the agreement for reduced nitrogen is reasonable. Large discrepancies between the models are found for oxidised nitrogen. The total deposition of NO_y in the EMEP model is about the same as in the OPS model, but the contribution from emissions in the Netherlands is much lower in the EMEP model. This difference can be traced back to the lower dry deposition and higher wet deposition of NO_y in the EMEP model compared with the OPS model. Consequently, in the EMEP model the deposition of NO_y is more a transboundary issue than in the OPS model.

Furthermore, the NO_x concentration in the EMEP is about 40% less than the measurements, indicating that a large fraction of the NO_x emissions is transported out of the Netherlands. We have shown that this can not be the result of a dry deposition velocity in the EMEP model that is too low, since the model also calculates a low dry deposition. It is also not likely that it is caused by sources outside Europe that are too large. Possible causes of the problems with NO_x/NO_y in the EMEP model might be that:

- The model does not properly take into account the contribution of low emission sources, e.g., cars, heating of houses.

- The limited vertical resolution of the model results in surface concentrations of NO_x that are too low. A surface correction term might be needed to convert the grid concentration to a surface concentration.

- There is a mismatch in the chemical balance of nitrogen species (NO_x, NO_y), e.g., too much HNO_3, PAN or other secondary species are produced, reducing the NO_x concentration.

- The contributions of the boundary and initial conditions are too large in the model, resulting in a too large contribution to NO_x concentrations from a long distance relative to local contributions.

A detailed study of all the nitrogen species in the EMEP model is necessary to resolve this issue.

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