Chapter 4

Source-receptor calculations

Peter Wind, David Simpson and Leonor Tarrasón

Over the past year a number of extensive investigations have been conducted, and production of source-receptor (SR) runs commenced for provision to IIASA under the EU CAFE-BASELINE project (Amann et al. 2004) and to the EU-MERLIN project (MERLIN 2001). This chapter provides an overview of the investigations undertaken during the last year, and of the new methodologies for SR matrices.

The data provided will be used for integrated assessment purposes and help in the determination of the response to emission changes. The SR tables provided in Appendix B give an overview over the main contributors to the transboundary air pollution for each country.

4.1 Introduction

Source-receptor (SR) matrices give the change in various pollutant indicators in each receptor country (or grid square) resulting from a change in anthropogenic emissions from each emitter country. Such matrices are generated for each country by reducing emissions for each country of one or more precursors by a given percentage, re-running the EMEP model, and comparing the resulting fields with the base-case or reference fields.

Our recent calculations have involved changes in emissions of $SO_x$, $NO_x$, $NH_3$, NMVOC, fine-primary particulates (PPMfine) and coarse primary particulates (PPM-coarse). The primary particulate emissions are assumed inert and so have no effect on the chemistry associated with the remaining four gaseous precursors. The other
pollutants show, however, significant interactions which must be accounted for.

In the appendix the most important SR matrices are presented. These tables and indeed the country-to-grid results from which they are constructed will be made available as ASCII files on the EMEP web-site, http://www.emep.int.

The results presented here are based on a series of scenario runs performed during the past year. These include:

- CLE SR runs for all the UNECE countries, S,N,A,V reductions (2003 meteorology)
- CLE SR runs for all the UNECE countries, S,N,A,V reductions (2000 meteorology)
- CLE SR runs for all the EU+ countries, S,N,A,V reductions (1997 meteorology)
- CLE SR runs for 11 emission sectors in the EU+ countries, S,N,A,V reductions (2003 meteorology)
- M50 SR runs for all the EU+ countries, A,N reductions (2003 meteorology)

For the definitions of CLE, M50, EU+ and S,N,A,V see section 4.2 below.

### 4.2 Notation

Table 4.1 summarises the notation used for emission precursors and pollutant indices throughout this chapter. With this notation, the change in any measure $X$ resulting from a change in emissions from country $i$ is given by $^1$:

$$
\Delta X_i = \left( \frac{\partial X}{\partial S_i} \right) \cdot \Delta S_i + \left( \frac{\partial X}{\partial N_i} \right) \cdot \Delta N_i + \left( \frac{\partial X}{\partial A_i} \right) \cdot \Delta A_i + \left( \frac{\partial X}{\partial V_i} \right) \cdot \Delta V_i \quad (4.1)
$$

For SR calculations, the derivatives were found by reducing the emissions by 15% of CLE, as this approximates to the mathematical derivative used above, but is sufficiently large to give a clear signal in the pollution changes. We will loosely use the notation $(\partial X/\partial E)$ to refer to the derivatives arising from the model’s SR calculations, and $\Delta X$ for the changes corresponding to arbitrary emission changes as given in eqn. 4.1.

$^1$Note that we here ignore the influence of primary particulate Matter, PPM, since these are inert and hence have no influence on sulphur-nitrogen-ozone chemistry in the current model.
CHAPTER 4. SOURCE-RECEPTOR CALCULATIONS

Table 4.1: General notation for emission precursors and pollutant changes used in this chapter

<table>
<thead>
<tr>
<th>Symbol</th>
<th>Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>$X$</td>
<td>Measure of pollution, e.g., concentration of $O_3$ or $PM_{2.5}$, AOT40, or deposition of sulphur ($S_{dep}$).</td>
</tr>
<tr>
<td>$N$</td>
<td>Emissions of $NO_x$</td>
</tr>
<tr>
<td>$S$</td>
<td>Emissions of $SO_x$</td>
</tr>
<tr>
<td>$A$</td>
<td>Emissions of Ammonia, $NH_3$</td>
</tr>
<tr>
<td>$V$</td>
<td>Emissions of VOC</td>
</tr>
<tr>
<td>$P$</td>
<td>Emissions of primary particulate matter, PPM</td>
</tr>
<tr>
<td>$E$</td>
<td>Emissions of any compound ($A, V, S, N$)</td>
</tr>
<tr>
<td>$\Delta X$</td>
<td>Change in $X$</td>
</tr>
<tr>
<td>$\Delta X_i$</td>
<td>Change in $X$ resulting from an emission change in country $i$</td>
</tr>
<tr>
<td>$\partial X$</td>
<td>Small change in $X$</td>
</tr>
</tbody>
</table>

Some deposition labels:
- OXS: Oxidised sulphur
- OXN: Oxidised nitrogen
- RDN: Reduced nitrogen

Base-cases and Deltas

The starting point for most SR calculations has been the so called Current Legislation Emissions (CLE) – those expected for the year 2010 according to current plans. These CLE emissions have been provided by IIASA (March 2004 delivery, see chapter 3 for details).

As part of the MERLIN project and preparation for the EU CAFE-BASELINE project, we have looked at a number of sensitivity tests which involve reduced base-cases for what we term EU$^+$, that is the 25 EU countries as well as Norway, Switzerland and Iceland. As well as CLE emissions, we look at a number of different ‘Base50’ base-cases for these countries, as given in Table 4.2. These cases have for simplicity been defined as simple 50% changes with respect to the CLE emissions. The locations of CLE, and of the base-cases A50 and N50, are illustrated in the two-dimensional example of Fig 4.1. This figure also illustrates the $\Delta E$ values for A and N emissions. From the CLE point (top right), we have $(\partial X/\partial N)_{CLE}$ and $(\partial X/\partial A)_{CLE}$. As noted above, for practical purposes we use emission changes of 15% of CLE in order to evaluate these differentials, and the same magnitude of change is applied even when starting from other base-cases. Thus, the changes applied from M50 amount to 15%
Table 4.2: Definition of bases cases in terms of emissions, as expressed relative to CLE 2010 values. CLE applies to all countries. All other reductions apply to 28 ‘EU+’ countries – see text.

<table>
<thead>
<tr>
<th>Base-case</th>
<th>S</th>
<th>N</th>
<th>A</th>
<th>V</th>
</tr>
</thead>
<tbody>
<tr>
<td>CLE</td>
<td>100</td>
<td>100</td>
<td>100</td>
<td>100</td>
</tr>
<tr>
<td>M50</td>
<td>50</td>
<td>50</td>
<td>50</td>
<td>50</td>
</tr>
<tr>
<td>S50</td>
<td>50</td>
<td>100</td>
<td>100</td>
<td>100</td>
</tr>
<tr>
<td>N50</td>
<td>100</td>
<td>50</td>
<td>100</td>
<td>100</td>
</tr>
<tr>
<td>A50</td>
<td>100</td>
<td>100</td>
<td>50</td>
<td>100</td>
</tr>
<tr>
<td>V50</td>
<td>100</td>
<td>100</td>
<td>100</td>
<td>50</td>
</tr>
</tbody>
</table>

of CLE emissions, or 30% of M50 emissions.

4.3 Previous investigations

SR relationships have been calculated for many years with the Lagrangian acidification and ozone models. For example, the first full set of such S-R matrices for ozone was presented in Simpson et al. (1997), where calculations were performed with the Lagrangian oxidant model for a 5 year period. A number of matrices with different assumptions (including 40% reductions) were calculated in order to supply results needed for the IIASA RAINS-ozone model (Schöpp et al. 1999).

The first set of matrices generated in this way with the EMEP Eulerian model was presented in Tarrasón et al. (2003). In this work the Eulerian model was run twice for each country. In one run, emissions of both S and N were reduced simultaneously. In the second run, emissions of A and V were reduced. It was hoped at the time that the emission-pairs S+N, and A+V, had sufficiently different effects that each run could be used to generate separate matrices for S, N, A, V. For example, since VOC mainly affects ozone and NH$_3$ mainly affects acidification, it was hoped that ozone changes could be entirely attributed to V and acidification changes entirely attributed to A. If true, this procedure would have generated SR matrices for 4 emission precursors at the cost of only two runs of the EMEP model per country.

Subsequent testing has show that this procedure would indeed work very well if the pollutants of interest were just O$_3$ (or AOTs) or depositions of sulphur, oxidised or reduced nitrogen. Unfortunately, it has been found that the effects of S and N are linked as far as their effects on particulate sulfate, nitrate and ammonium are concerned. Even A and V were found to have significant inter-relationships so that these two precursors should also be treated separately.
Consequently, the methodology to calculate source-receptor relationships has changed and, at the time of writing, there seems little choice but to run the EMEP model separately for all 4 precursor emissions.

### 4.4 Investigations of SR relationships

For small changes equation 4.1 is a correct approximation. In practice, to find the value of the derivatives, we make a run with emissions slightly perturbed and calculate how much the quantity $X$ varies. In principle we need a separate run for each precursor: four runs per country (S,N,A,V). Four runs are enough because PPM do not interact with the other components, hence we can calculate the P runs simultaneously with any of the others. We have typically run N+P together.

However, as we move away from the base-case used to obtain these derivatives,
this equation will start to fail, because there are mixed terms (higher order derivatives) in the exact equation. New sets of derivatives should ideally be calculated for a large number of base-cases, but there are too many possible emission regimes to allow this to be done in practice. Instead, we aim to find how the derivatives change with respect to emission regime. Hopefully, some derivatives are sufficiently constant, or predictable in simple terms, that only a few extra runs of the model are needed in order to generate general source-receptor relationships.

Over the past year a number of investigations have been conducted in order to learn about the important factors affecting SR calculations and their use. These include:

NONLIN test runs - Continuation of last years investigations to determine the new methodology to calculate SR relationships.

Mass balance tests - Evaluation of the mass conservation properties of the deposition results to recommend methods for presentation of source-receptor matrices for depositions

Linearity tests - Evaluation of the linearity of the results for air concentrations and indicators and analysis of the ability of equation 4.1 to reproduce results for relatively large emission change is tested.

Base50 tests - Investigation of the way in which \(\frac{\partial X}{\partial E}\) and SR matrices change depending on the base-case or reference emission regime from which they are calculated.

Some of the findings of these investigations are presented below:

### 4.5 NONLIN tests

In the project “Preparatory Actions in the Public Health and Environmental Policy Sector-Lot 10 Air Pollution”, 2004 (also called NONLIN) detailed linearity tests have been performed for four countries (Italy, Netherlands, Germany and United Kingdom). Runs starting from three different emissions levels (CLE, MFR (Maximum Feasible Reduction) and UFR (Ultimate Feasible Reduction = MFR/2)) and seven types of reduction scenarios (N, S, A, V, S+N, A+V, N+S+A+V) have been compared. We will just give the main conclusions here.

An important question that this study addressed was the possibility of combining pollutants together (S+N and A+V) in the emission reduction scenarios, without losing to much accuracy. This would be possible if at least two of the terms in equation 4.1 are sufficiently small, which means that all the pollutants \(X\) are influenced by only one of the emitted pollutant (S or N) and (A or V) at a time. However some components, specially Nitrate and Ammonium air concentrations showed a dependency on all the four emitted pollutants S, N, A and V.

This study concluded therefore that for integrated assessment purposes the four different pollutants S, N, A, V should be considered separately.
4.6 Mass balance investigations

An emission reduction will affect the pollutants in two qualitatively different ways:

1. A removal of the pollutants which contained the same chemical elements as removed from the emissions.

2. A modification of the composition of the atmosphere which can lead to a change in the chemistry and deposition rates.

The first effect is the simplest to understand. For example for PPM this is the only effect present and it is therefore easy to trace the sources. For depositions of OXS, OXN or RDN the second effect is present but generally smaller than the first one. For \( \text{O}_3 \) the first effect is absent and \( \text{O}_3 \) is only affected by changes in the chemical composition of the atmosphere. A modification of the composition of the atmosphere will also lead to nonlinear effects which are more difficult to describe and quantify.

There are two conservation properties which have been tested:

1. Can all pollutants emitted by a country be accounted for?

2. Can all the pollutants deposited at a given location (a country, a region or an individual gridcell) be traced back to their source?

4.6.1 Mass conservation of emitted pollutants

It is important to verify that all pollutants emitted by a country can be accounted for. Most of the emitted pollutants will end up as deposition. The result obtained by summing the depositions from a country to all areas, can be compared to the total amount emitted by that country. For a complete picture we should also take into account the amount of pollutants that cross the lateral or top boundaries of the model domain. Also the total amount of pollutants in the atmosphere at the beginning and end of the run should be included in the budget:

\[
X_{\text{EMISSIONS}} + X_{\text{INFLUX}} + X_{\text{INITIAL AIR CONC}} = X_{\text{DEPOSITION}} + X_{\text{OUTFLUX}} + X_{\text{END AIR CONC}}
\]

where \( X \) is any chemical element in the calculations. This is the mass conservation law and it applies even if non-linear effects are present. Tests with the Unified EMEP model have already shown that the model is mass conservative.

An example of this mass conservation can be found for the depositions of oxidised sulphur, oxidised nitrogen and reduced nitrogen. For countries far from the domain boundaries the in and out fluxes \( X_{\text{INFLUX}} \) and \( X_{\text{OUTFLUX}} \) are negligible or small. For yearly averages (as presented in this report), the quantity of pollutants in the air at the start and end of the run \( X_{\text{INITIAL AIR CONC}} \) and \( X_{\text{END AIR CONC}} \) are also negligible compared to the other terms. The results given in the SR tables (see Appendix B)
confirm that the amount of pollutant emitted by a given country, and the sum of the pollutants deposited originating from that country (sum following a column) are usually close to each other. For countries closer to the boundaries the quantity of pollutants emitted can be larger than the corresponding depositions. For RDN deposition the differences are generally smaller than for OXN and OXS, because RDN has a shorter lifetime and shorter residence times in the atmosphere and is therefore less likely to leave the model domain.

4.6.2 Budget of deposited pollutants

An SR matrix distributes the responsibility for the pollutants. The total amount of pollutants deposited at a given location is expected to be attributed unequivocally to identified sources. For every gridcell the sum of the contributions from every single source should then be equal to the total amount of deposited pollutant. In practice, it is not always possible to define a unique cause, or assign the responsibility for the amount deposited, and the totals may differ from the amount deposited.

As an example, a component like \( \text{SO}_x \) emitted by one country can combine with \( \text{NH}_4 \) originating from a second country to form ammonium sulfate before it is deposited in a third country. In this situation we can follow the Sulphur atom and assign the Sulphur deposited to the country which has emitted the Sulphur atom. However ammonium sulfate has a very different transport pattern than \( \text{SO}_x \). Therefore the effect of \( \text{NH}_4 \) will be to change the deposition map of \( \text{SO}_x \), even if the total amount of Sulphur deposited is unchanged. In this context the emission of \( \text{NH}_3 \) from one country can contribute to the deposition of Sulphur in another country, also when the sulphur originates from a third country.

For \( X = \text{Deposition of Oxidised Sulphur} \) in equation 4.1, the first derivative is normally much larger than the other three, representing the rate of variation of OXS deposition due to \( \text{SO}_x \) emissions changes. This term will usually be positive indicating that a reduction of \( \text{SO}_x \) emissions usually results in a reduction of OXS deposition. The three other terms represent the effect of respectively \( \text{NO}_x \), \( \text{NH}_3 \) and VOC on OXS depositions. Since a change of \( \text{NH}_3 \) emission for instance does not change the total amount of Sulphur present (either in the atmosphere or deposited), this term will be positive at some locations and negative at others, depending on the ratio between \( \text{NH}_3 \) and \( \text{SO}_2 \) and its effect on dry deposition and chemical reactions. These terms will have an influence on where the \( \text{SO}_x \) will be deposited, but not on the total amount deposited.

Thorough testing of the model has shown that for depositions, there is a very strong linearity. We have therefore extrapolated the results obtained from our 15% emission reductions to generate equivalent results for 100% emission reductions, assuming complete linearity. These 100% results are in fact those presented in the Appendix B for deposited pollutants.

Figure 4.2 shows two maps with the relative differences between the amount of OXS deposited and the sum of the individual contributions from every \( \text{SO}_x \) emitter (i.e. each UNECE country + all other sources. This corresponds to sums following
rows in the SR tables):
\[
\frac{(\sum_{all} \Delta X_i) - \Delta X_{all}}{\Delta X_{all}}
\]

The figures show the relative differences in two cases: the figures on the left are computed using all terms from equation 4.1, while the figures on the right show results obtained using only the largest term. The corresponding maps for the deposition of oxidised nitrogen and reduced nitrogen are shown in Figures 4.3 and 4.4. These maps allow us to quantify the ability of the SR matrices to unequivocally assign the responsibility for the different pollutants.

Positive values show areas where part of the deposited pollutant has not been accounted for, whereas negative values indicate that the sum of country-specific depositions is greater than the modelled total deposition. Given the extrapolation from SR matrices derived from 15% emission changes to 100% emission changes assumed here, then full 100% agreement would be surprising.

Figure 4.2: Fractional difference between the total deposition of oxidised Sulphur in the base case and the sum of all countries/region contributions. Left figure: the individual contributions are calculated as the sum of the contribution due to S, N, A and V reductions. Right figure: the individual contributions are calculated using only the contribution due to S reductions. (Unit: fraction)

For OXS deposition, the relative differences between total deposition and sum of emitter contributions are small (less than 3% almost everywhere). This shows that the SR has assigned an unique origin to all the deposited Sulphur. The small differences between the left and right figure 4.2 show that the redistribution of pollutants caused by NOx, NH3 and VOC, play a secondary role, even after the extreme scaling we have done from 15% to 100% emission reductions. It is interesting to note that the values
Figure 4.3: Fractional difference between the total deposition of oxidised Nitrogen in the base case and the sum of all countries/region contributions. Left figure: the individual contributions are calculated as the sum of the contribution due to S, N, A and V reductions. Right figure: the individual contributions are calculated using only the contribution due to N reductions. (Unit: fraction)

are smaller in the case where all terms from equation 4.1 are included (left figure 4.2) than if only the dominant term is taken into account (right figure 4.2).

For oxidised and reduced N there are some small areas where the differences reach more than 10%. Differences are larger for nitrogen than for sulphur depositions because both ammonia and nitrogen oxides play an important role in the chemistry and transport of oxidised and reduced nitrogen. As it could be expected, the differences between the total run and the sum of all countries and regions are larger when we consider only N or A runs than when we consider the contribution from all pollutants.

The overall conclusions from this test are that:

- The sum of all four emission derivatives should be included when analysing the depositions of sulphur and nitrogen compounds. This is the reason for including all the four components in the depositions SR relationships presented in Appendix B.

- The behaviour of depositions is very linear, even after extrapolating results derived from 15% emission reductions to 100% reductions. This suggests that deposition SR generated by the EMEP model can to some extent be considered as entirely linear.
4.7 Linearity test: EU25 and M50

For sufficiently large values of $\Delta E$, equation 4.1 is expected to become less accurate because nonlinear effects can no longer be neglected. In order to test the applicability of this equation for large emission changes we have compared the results obtained using equation 4.1 with the results obtained by direct calculation (i.e. including all nonlinear effects).

4.7.1 EU25

In this test we consider a 15% reduction of emissions for one component (S, N, V or A) simultaneously for all 25 EU countries starting from CLE. The changes (denoted $\Delta X_{EU25}$) obtained by this run can then be compared with the sum of the corresponding contributions from the 25 individual countries (denoted $\sum_{i=1}^{25} \Delta X_i$). To simplify the discussion below we will loosely use the term $\Delta X/\Delta E$ to denote changes $\Delta X$ caused by emission changes $\Delta E$, even though we are not actually dividing by the emissions.

Figure 4.5 show the ratio of changes obtained by the two methods. For $\Delta SO_4/\Delta S$, $\Delta aNO_3/\Delta N$, $\Delta NH_4/\Delta A$, $\Delta DDEP_{OXN}/\Delta N$ and $\Delta DDEP_{RDN}/\Delta A$ the differences between the two methods are below 5%. (Except for points where the changes are extremely small, and the ratio becomes meaningless). $\Delta AOT40/\Delta N$ can have both positive and negative sign around the North Sea, giving an unstable ratio. (We here
The overall result from this test is that the consequences of an emission change around 15% by several countries can be obtained by simply adding the changes caused by the individual countries. At this level the nonlinear effects can certainly be neglected.

4.7.2 M50

As a much stricter test of the applicability of the SR results, we here compare two estimates of pollution levels for the M50 base-case, one obtained by extrapolation of CLE results to M50 levels using the SR derivatives and eqn 4.1, and the other obtained by a direct run of the EMEP model for M50 emissions. The M50 emissions level was discussed in section 4.2 and consists of a 50% emission reduction of S, N, A and V, for 28 countries relative to CLE. This emission level will considerably modify the chemical composition of the atmosphere.

From the direct estimate, we simply obtained the difference \( \Delta X_{M50} = X(\text{CLE}) - X(M50) \) from two model runs, where \( X(\text{CLE}) \) and \( X(M50) \) are the values of \( X \) at CLE and M50 emission levels, respectively. For the SR-derived estimate, we extrapolate the CLE results down to the M50 level using the \( \Delta X_i \) values obtained in the SR calculations, which are based upon 15% emission reductions. This involves the sum of 112 (4 pollutants \( \times \) 28 countries) individual contributions. For any pollutant \( X \) we denote the change in \( X \) resulting from the sum of all of these changes as \( \Delta X_{\text{sumCLE}} \), where:

\[
\Delta X_{\text{sumCLE}} = \frac{50}{15} \sum_{i=1}^{28} \Delta X_i
\]

Figure 4.6 present the ratios \( \Delta X_{\text{sumCLE}}/\Delta X_{M50} \) for \( X = \text{AOT40}, \text{SO}_4, \text{aNO}_3, \text{NH}_4, \text{DDEP}_\text{OXN} \) or \( \text{DDEP}_\text{RDN} \).

The AOT40 levels show a complex pattern, with both positive and negative differences. For \( \text{SO}_4 \) concentrations the extrapolated results are mostly within 5% of the M50 results. The concentrations of \( \text{aNO}_3 \) and \( \text{NH}_4 \) differ in the southern part of Europe with differences of the order of 10-20%. For dry deposition of OXN over land the differences are of about 20%. The dry deposition of RDN over land areas show only small differences (5%).

Even if there are non-negligible differences between the two sets of results, this test shows that the CLE results can be extrapolated to relatively large emission changes without becoming meaningless.
Figure 4.5: Ratio of the changes ($\Delta X$) obtained by the sum of changes from 25 individual country runs divided by the changes obtained with all EU countries emissions reduced together, i.e. of $\sum_{i=1}^{25} \Delta X_i / \Delta X_{EU25}$. Changes labelled $dX/dE$ actually represent ratios of $\Delta X$ caused by change in emissions of $E$. (units: ratio in %)
Figure 4.6: Ratio between the sum of the changes obtained by application of the SR derivatives over a 50% emission reduction for 28 individual EU countries compared to direct calculation of the change with simultaneous country emission reductions, i.e. of \( \frac{\sum_{i=1}^{28} \Delta X_i}{\Delta X_{M50}} \) (see text). Results shown for \( \Delta AOT40, \Delta SO_4, \Delta NO_3, \Delta NH_4, \Delta DDEP\_OXN \) and \( \Delta DDEP\_RDN \). (units: %)
4.8  Base50 tests: Variation of $\frac{\partial X}{\partial E}$

In order to understand the sensitivity of the various $\frac{\partial X}{\partial E}$ derivatives to the base-case emissions chosen for their calculation, a number of sensitivity tests have been conducted to calculate these values at different points. As well as the CLE 2010 base case, 5 other base-cases were defined in which emissions of either S, N, A or V (or all) were reduced by 50% for the EU$^+$ countries (Table 4.2). (These tests have focused on EU$^+$ and western-central Europe in order to meet the tight deadlines imposed by the EU-funded MERLIN and CAFE-BASELINE projects.)

In view of the very large number of scenarios to be explored, full-year runs of the model were not possible. Instead, the model has been run for January and July (of 2003) and a pseudo-yearly average derived from this.

It should be noted that the cases S50, N50, A50 and V50 (c.f. Table 4.2) are extreme, as it is very unlikely that future emission scenarios involve cases where emissions of only one pollutant are reduced while the others remain at 100% of CLE levels. The M50 base-case is chosen to represent a very simple but quite plausible future emission scenario where all emissions are reduced by 50%. However, where the M50 derivatives are very different from the CLE ones, the extreme "Base50" cases serve to illustrate which of the emission reductions associated with M50 is actually most responsible for the change in $\frac{\partial X}{\partial E}$.

Thus, the main aims of these tests are to (1) establish how different derivatives derived at CLE are compared to those derived at M50, and (2) if different, which emission changes in the base-cases are most responsible for the differences.

For each of these base-cases, $\frac{\partial X}{\partial E}$ values were calculated for two example countries: Great Britain or Hungary. These countries have been chosen to represent one country with high-emissions (GB), particularly of NO$_x$, and one with low emissions (HU). With 2 emitter countries (GB, HU), 4 emitter-pollutants (S,N,A,V) and 6 base-cases, we have a total of 48 scenario results to analyze for each pollutant of interest. In order to derive simple numbers for comparison, changes in each pollutant X were summed across all land-based grid squares within a rectangular region of the EMEP grid covering western-central Europe (EMEP x=35-105,y=1-89). Further, absolute values of changes were accumulated, so that both negative and positive $\Delta X$ values were summed equally, in order to avoid situations where the effects of a country’s emissions were found to be low because of cancellation of $\Delta X$ values of different signs. These summed values are denoted ‘Sum $|\Delta X|$’.

These summary results are presented in Table 4.3, where each column of numbers gives the change in Sum $|\Delta X|$ arising from the emission reduction of S, N, A or V from (a) GB and (b) HU. Although these Sums have not been divided by the appropriate emissions or grid area, they are proportional to the related $\frac{\partial X}{\partial E}$ values and for simplicity we will refer to the latter notation in the discussions below.
Table 4.3: Variation of changes in \( \text{Sum} \Delta X \) (see text) with base-case, for emissions reductions of S, N, A or V in (a) Great Britain and (b) Hungary. Units are omitted for clarity.

| Pollutant (X) | Base-case | \( \text{Sum} |\Delta X| \) (a) : emitter GB | \( \text{Sum} |\Delta X| \) (b) : emitter HU |
|---------------|-----------|-----------------|-----------------|
|               | Emission reductions: S N A V | S N A V | S N A V |
| O_3           | CLE       | 3.58 50.81 0.59 | 76.86 | 0.65 | 13.52 | 0.13 | 5.58 |
|               | M50       | 5.53 47.36 0.91 | 70.35 | 1.13 | 19.55 | 0.17 | 5.35 |
|               | S50       | 6.47 50.86 0.65 | 77.36 | 1.74 | 13.55 | 0.08 | 5.59 |
|               | A50       | 3.60 50.74 1.31 | 76.94 | 0.63 | 13.53 | 0.25 | 5.58 |
|               | N50       | 3.49 54.87 0.43 | 55.87 | 0.60 | 20.48 | 0.13 | 4.84 |
|               | V50       | 3.25 58.88 0.55 | 89.64 | 0.55 | 13.01 | 0.12 | 5.45 |
| AOT40         | CLE       | 0.63 4.15 0.09 | 14.23 | 0.36 | 7.05 | 0.04 | 0.57 |
|               | M50       | 0.51 4.59 0.04 | 6.61 | 0.47 | 6.82 | 0.04 | 0.37 |
|               | S50       | 1.19 4.25 0.10 | 14.53 | 0.94 | 7.15 | 0.02 | 0.59 |
|               | A50       | 0.65 4.19 0.15 | 14.29 | 0.35 | 7.06 | 0.09 | 0.57 |
|               | N50       | 0.41 8.83 0.04 | 6.85 | 0.26 | 7.60 | 0.02 | 0.31 |
|               | V50       | 0.45 2.63 0.06 | 12.14 | 0.30 | 6.39 | 0.03 | 0.63 |
| SO_4          | CLE       | 6.25 0.95 0.26 | 0.99 | 13.10 | 0.48 | 0.20 | 0.11 |
|               | M50       | 6.62 0.44 0.53 | 0.71 | 13.34 | 0.40 | 0.20 | 0.10 |
|               | S50       | 6.25 0.69 0.13 | 0.76 | 13.29 | 0.30 | 0.12 | 0.09 |
|               | A50       | 6.64 1.00 0.96 | 1.02 | 13.12 | 0.48 | 0.21 | 0.11 |
|               | N50       | 6.31 0.59 0.28 | 0.61 | 13.20 | 0.75 | 0.21 | 0.10 |
|               | V50       | 6.18 1.20 0.26 | 1.30 | 12.89 | 0.47 | 0.20 | 0.11 |
| aNO_3         | CLE       | 0.78 3.71 2.73 | 0.32 | 2.50 | 1.48 | 3.02 | 0.06 |
| (fine)        | M50       | 0.59 2.60 1.72 | 0.14 | 2.10 | 1.67 | 3.24 | 0.04 |
|               | S50       | 0.88 4.03 2.74 | 0.35 | 2.85 | 1.67 | 2.80 | 0.06 |
|               | A50       | 0.58 1.70 2.75 | 0.19 | 2.00 | 0.99 | 4.40 | 0.05 |
|               | N50       | 0.51 4.67 1.50 | 0.18 | 1.80 | 2.08 | 1.89 | 0.04 |
|               | V50       | 0.78 3.54 2.71 | 0.45 | 2.39 | 1.38 | 2.90 | 0.06 |
| NH_4          | CLE       | 4.45 2.96 2.83 | 0.98 | 5.87 | 1.43 | 4.50 | 0.11 |
|               | M50       | 4.82 2.39 2.22 | 0.56 | 6.63 | 1.77 | 5.05 | 0.08 |
|               | S50       | 4.75 3.47 2.81 | 0.84 | 7.24 | 1.57 | 3.35 | 0.11 |
|               | A50       | 4.20 1.08 3.81 | 0.80 | 4.51 | 0.94 | 8.54 | 0.08 |
|               | N50       | 4.75 4.83 1.61 | 0.45 | 6.50 | 2.48 | 3.42 | 0.08 |
|               | V50       | 4.41 2.60 2.81 | 1.40 | 5.89 | 1.35 | 4.37 | 0.11 |
| pNO_3         | CLE       | 0.19 1.92 0.32 | 0.19 | 0.55 | 0.57 | 0.58 | 0.02 |
| (coarse)      | M50       | 0.16 2.16 0.20 | 0.18 | 0.53 | 0.62 | 0.62 | 0.01 |
|               | S50       | 0.25 1.86 0.34 | 0.18 | 0.71 | 0.52 | 0.57 | 0.01 |
|               | A50       | 0.16 2.10 0.28 | 0.20 | 0.43 | 0.65 | 0.79 | 0.02 |
|               | N50       | 0.12 2.00 0.18 | 0.15 | 0.40 | 0.61 | 0.38 | 0.01 |
|               | V50       | 0.19 1.90 0.33 | 0.20 | 0.53 | 0.53 | 0.56 | 0.02 |
4.8.1 Ozone, $\partial O_3/\partial E$

Table 4.3 clearly shows that for all base-cases, it is the N and V emissions which dominate changes in $O_3$. For GB emissions the the values of $\partial O_3/\partial N$ and $\partial O_3/\partial V$ are similar in the M50 base-case to the CLE case, whereas for HU emissions the value of $\partial O_3/\partial N$ is significantly higher in M50 than CLE. Examination of the other Base50 cases shows that the N50 case is similar, implying that the lower NOx emissions in M50 cause this different values of $\partial O_3/\partial N$.

Of the extreme "Base50" cases, it is the N50 scenario which seems to be most important in explaining deviations between CLE and M50 for both countries. $\partial O_3/\partial N$ is found to be not very sensitive to N50 emissions levels for the high-NOx GB case, but sensitive in the low-NOx HU case. $\partial O_3/\partial V$ is most sensitive to N50 levels for GB and HU, and sensitive also to V50 for the GB case. These findings are consistent with those found previously for ozone sensitivities in Europe (Simpson 1991, 1995, Simpson and Malik 1996, Simpson et al. 1997) and justify why the SR tables for ozone concentrations are presented both for NOx and VOC emission reductions.

4.8.2 AOT40^3m, $\partial\text{AOT40} / \partial E$

Figure 4.7 shows the values of AOT40^3m (from one month only in this case, July) for both the CLE and M50 cases. Both cases clearly show AOT40 maxima over southern Europe and especially Italy for this month. Peak values of well over 3000 (arbitrary units) in the CLE case are reduced to just around 2000 as a result of the emission reductions.

Figure 4.8 presents some results of the tests where N emissions from the United Kingdom are reduced. Figure 4.8(left) illustrates the actual reductions in AOT40 ($\Delta\text{AOT40}$) found when GB emissions are reduced 15% from CLE levels. (Note, $\Delta\text{AOT40}$ is positive when AOT40 levels are reduced by the N-emissions reduction). As frequently occurs for ozone and ozone-based indicators, both positive and negative changes are observed. In south-east England, where NOx levels are highest, reduction in NOx emissions causes an increase in AOT40 levels, whereas over most of Europe AOT40 levels are reduced by this emission change.

Figure 4.8(b) illustrates the ratio of the values of $\partial\text{AOT40} / \partial N$ obtained in the M50 test-case compared to the value of $\partial\text{AOT40} / \partial N$ obtained in the CLE case, expressed as a percentage. Thus, the light green shades indicate areas where the derivative is similar in the Base50 case to the CLE case.

For M50, Figure 4.8(b) shows that over much of the United Kingdom itself the value of $\partial\text{AOT40} / \partial N$ is actually much smaller in the M50 case compared to the CLE values - the ratio is often near zero %. Over most of Europe, however, the ratio is much closer to 100%, and increases further over Italy and parts of the Mediterranean. Close inspection reveals that large percentage deviations from 100% are usually associated with areas where either the M50 values of AOT40 had become low (e.g. over GB itself, c.f. Fig. 4.7), or where the $\Delta$ AOT40 values are low (c.f. Fig 4.8a).
Figure 4.7: Calculated values of AOT40 from JAN+JUL runs (here only July applies), 2003 meteorology, with (a) CLE 2010 emissions (left) and (b) Base-M50 emission (right). Note possibly different scales. Arbitrary units.

Figure 4.9 presents similar information but now with emissions reduced in Hungary. In this case the $\partial \text{AOT40}/\partial N$ values are similar in both the M50 and CLE cases – ratios are close to 100%, especially in areas where the CLE $\partial \text{AOT40}/\partial N$ values are significant. (Indeed, although not shown, the ratio is close to 100% for all base-cases).

As a final illustration for AOT40, Figure 4.10 illustrates changes in the $\partial \text{AOT40}/\partial V$. M50 values are somewhat lower than CLE though, being most influenced by the N50 case (not shown). For the equivalent Hungarian case (not shown), the values of $\partial \text{AOT40}/\partial V$ in the Base50 tests are even closer to 100% of CLE values.

Table 4.3 further shows that for all base-cases, it is the N and V emissions which dominate changes in AOT40. Further, the overall (summed) values of $\partial \text{AOT40}/\partial N$ are similar across most base-cases, and most importantly between CLE and M50. One exception is that the values of $\partial \text{AOT40}/\partial V$ are much lower in the M50 case than in the CLE case, by more than a factor of 2 for GB. The N50 test shows that this is due to the reduced nitrogen levels associated with M50.

In general, the derivatives are much more sensitive to the N50 and V50 base-cases than those of $O_3$, especially for GB emissions.

4.8.3 Sulfate, $\partial \text{SO}_4/\partial E$

Table 4.3 shows that for all base-cases it is changes in S emissions which dominate $\partial \text{SO}_4/\partial E$ values. The derivatives for S are very similar across all base-cases. These
Figure 4.8: Variability of $\Delta$AOT40/$\Delta N$ Derivatives (July only). Left figure shows $\Delta$AOT40, for the CLE case, when $N$ emissions from country GB are reduced by an amount equivalent to 15% CLE. Right figure shows the ratio of $\Delta$AOT40/$\Delta N$ values for the base-case M50 compared to the CLE case, expressed as a percentage. (Note possibly different scales for positive and negative values in left fig. Units are arbitrary for fig (a), and % for fig (b))

Figure 4.9: Variability of $\Delta$AOT40/$\Delta N$ Derivatives (July only). Left figure shows $\Delta$AOT40, for the CLE case, when $N$ emissions from country HU are reduced by an amount equivalent to 15% CLE. Right figure shows the ratio of $\Delta$AOT40/$\Delta N$ values for the base-case M50 compared to the CLE case, expressed as a percentage. (Note possibly different scales for positive and negative values in left fig. Units are arbitrary for fig (a), and % for fig (b))

findings are very consistent with those illustrated previously in section 4.7. An important conclusion is that to a good approximation $\partial SO_4/\partial E$ calculated at CLE can be applied to other emission regimes.
Figure 4.10: Variability of AOT40/ΔV Derivatives (July only). Left figure shows AOT40, for the CLE case, when V emissions from country GB are reduced by an amount equivalent to 15% CLE. Right figure shows the ratio of AOT40/ΔV values for the base-case M50 compared to the CLE case, expressed as a percentage. (Note possibly different scales for positive and negative values in left fig. Units are arbitrary for fig (a), and % for fig (b))

Figure 4.11: Variability of SO4/ΔS Derivatives (Jan+July). Left figure shows SO4, for the CLE case, when S emissions from country GB are reduced by an amount equivalent to 15% CLE. Right figure shows the ratio of SO4/ΔS values for the base-case M50 compared to the CLE case, expressed as a percentage. (Note possibly different scales for positive and negative values in left fig. Units are arbitrary for fig (a), and % for fig (b))

Figure 4.11 illustrates the simplicity of the sulphur system with respect to United Kingdom emissions. The $\partial$SO4/$\partial$S values obtained in M50 (and although not shown, all base-cases) are close to those of the CLE case (i.e. the ratios shown are close to
Figure 4.12: Variability of $\Delta a\text{NO}_3/\Delta N$ (fine-nitrate) Derivatives (Jan+July). Left figure shows $\Delta a\text{NO}_3$, for the CLE case, when $N$ emissions from country GB are reduced by an amount equivalent to 15% CLE. Right figure shows the ratio of $\Delta a\text{NO}_3/\Delta N$ values for the base-case M50 compared to the CLE case, expressed as a percentage. (Note possibly different scales for positive and negative values in left fig. Units are arbitrary for fig (a), and % for fig (b))

100%), and no serious deviations are visible in these figures.

4.8.4 Nitrate, fine-mode, $\partial a\text{NO}_3/\partial E$

Concentrations of fine particulate nitrate are sensitive to three precursors, S, N and A, with only V emissions having little effect (Table 4.3). These results suggest that at least three pollutant derivatives may be needed when analysing source-receptor relationships for PM. The M50 derivatives are somewhat different to the CLE derivatives, and most affected by the A50 and N50 base-cases. There is clearly a strong coupling between A and N, since $\partial a\text{NO}_3/\partial A$ is mainly influenced by the N50 base-case, whereas $\partial a\text{NO}_3/\partial N$ is mainly influenced by the A50 base-case.

As can be seen in Figures 4.12–4.13, some variation can be seen in the behaviour of the derivatives in different parts of Europe, but in general M50 values of $\partial a\text{NO}_3/\partial N$ vary between ca. 60-140% of CLE values. However, in those regions where $\Delta a\text{NO}_3$ values are significant (left sub-figures), then values are still close to 100%. Figure 4.14 shows the equivalent figure for reductions in A emissions for HU. Here again, M50 values of $\partial a\text{NO}_3/\partial S$ are quite similar to CLE values, although in the extreme S50 and especially A50 cases (not shown) greater discrepancies are seen.

4.8.5 Ammonium, $\partial \text{NH}_4/\partial E$

As with fine-mode nitrate above, concentrations of particulate ammonium ($\text{NH}_4\text{NO}_3$) are sensitive to the three precursors, S, N and A, with only V emissions having lesser
Figure 4.13: Variability of $\Delta a\text{NO}_3/\Delta N$ (fine-nitrate) Derivatives (Jan+July). Left figure shows $\Delta a\text{NO}_3$, for the CLE case, when $N$ emissions from country HU are reduced by an amount equivalent to 15% CLE. Right figure shows the ratio of $\Delta a\text{NO}_3/\Delta N$ values for the base-case M50 compared to the CLE case, expressed as a percentage. (Note possibly different scales for positive and negative values in left fig. Units are arbitrary for fig (a), and % for fig (b)).

Figure 4.14: Variability of $\Delta a\text{NO}_3/\Delta A$ (fine-nitrate) Derivatives (Jan+July). Left figure shows $\Delta a\text{NO}_3$, for the CLE case, when $A$ emissions from country HU are reduced by an amount equivalent to 15% CLE. Right figure shows the ratio of $\Delta a\text{NO}_3/\Delta A$ values for the base-case M50 compared to the CLE case, expressed as a percentage. (Note possibly different scales for positive and negative values in left fig. Units are arbitrary for fig (a), and % for fig (b)).

effects (Table 4.3). The M50 derivatives are somewhat different to the CLE derivatives, and most affected by the A50 and N50 base-cases. The same strong coupling between $A$ and $N$ exists, since $\partial \text{NH}_4/\partial A$ is mainly influenced by the N50 base-case, whereas
\( \partial \text{NH}_4 / \partial N \) is mainly influenced by the A50 base-case.

### 4.8.6 Nitrate, coarse-mode, \( \partial \text{pNO}_3 / \partial E \)

Coarse-mode nitrate is seen to be dominated by N emissions for the GB case, and by S, N, and A emissions for the HU case (Table 4.3). Derivatives are very similar across all the base-cases though, so to a good approximation \( \partial \text{pNO}_3 / \partial E \) calculated at CLE can be applied to other emission regimes.

### 4.8.7 Summary of Base50 tests

Table 4.4 summarises the above observations by indicating which parameters are most important for each pollutant X, and which of the Base50 base-cases the derivatives (\( \partial X / \partial E \)) are most sensitive to. For example, Table 4.4 indicates that the derivatives for O\(_3\) are sensitive to N and V emissions, and especially to the N50 levels among the base-cases. (The sensitivity to M50 is implied and not shown explicitly here, since M50 includes emissions in N50). Thus, it may be supposed that derivatives for O\(_3\)
calculated at CLE may need to be adjusted to reflect changes in nitrogen and VOC emissions associated with emission strategies, but that to a first approximation changes in any other emissions can be ignored.

Looking at all pollutants, this table suggests that values of $\partial X/\partial E$ are affected significantly by the changes in N-level and A-level when moving from CLE base-cases to other regimes. None of the pollutants were sensitive to the level of S emissions.

It should be stressed that the Base50 tests discussed above give only a general picture of the changes associated with these tests and their complexity. Results found for individual grid squares may well differ from those suggested above. However, although limited to runs with Jan+July only, and to 2 emission countries, the results of these tests seem to correspond well to other results obtained with full-year runs. A general finding is that model derivatives obtained at CLE emission levels may need to be modified for other emission regimes, and the main modifying factors are associated with the N and A levels. These results and many other need to be examined in more detail before specific recommendations can be made concerning applications of such derivatives in integrated assessment models, but they also suggest that in most cases the changes in derivatives are relatively modest.

As a result of these findings, the EMEP model results provided to IIASA and MERLIN have included derivatives calculated at CLE level, and also at M50. The latter calculations have so far been for $\partial X/\partial N$ and $\partial X/\partial A$ only, in order to conserve computer-time, whilst providing those derivatives which are most likely to change in moving from CLE to M50.

### 4.9 SR results for Appendix

All the results presented in Appendix B are based on model runs using meteorological input data for year 2000. The emissions are the CLE 2010 emissions (see chapter 3). For each country four separate runs have been performed: with an emission reduction of 15% for $\text{SO}_x$, $\text{NO}_x$(+PPM), $\text{NH}_3$ or VOC respectively.

It is important to note that the meteorological input from a different year would lead to considerable changes in most of the results, especially for the long-range transported pollutants.

The deposition tables show the results of these (15%) model runs after scaling with a factor 100/15, giving the equivalent of 100% emission reductions. Although introducing some small errors due to non-linearity (see section 4.6.2), this procedure allows an estimate of the complete deposition budget over each country. The depositions tables can thus be interpreted as the contributions from one country to another. (With the limitations discussed in 4.6.2)

For the concentrations and indicator tables, the differences obtained by the 15% emission reduction are given directly. The tables should be interpreted as the predictions of this reduction scenario.
These tables also include the changes in concentrations/depositions caused by changes (also of 15%) in:

**NAT** Natural+other sources, includes natural marine, volcanic and lightning emissions as well as aircraft.

**BIC** includes all boundary and initial components except O$_3$.

Ozone is excluded from BIC for these runs because of its special importance and known high contribution to indices such as AOT40. The role of background O$_3$ will be addressed in a separate study. BIC as used here includes anyway compounds which have undergone important changes due to man-made emissions, such as NO$_x$, CH$_4$ and CO (see chapter 6, Simpson et al. 2003).

### 4.10 SR results for country reports

In order to allow a concise and detailed summary of EMEP results for each country, 44 country-specific data reports have been prepared to accompany this main report (Klein et al. 2004). The tables and figures in these country reports are based on the same data as presented above and in the Appendix. The main difference is that the SR relationships are scaled to 2002 emissions for the country-reports, whereas the original SR computations used 2010 CLE emissions. The scaling of each concentration or deposition $X$ to 2002 emissions has been done by multiplying the changes of $X$ due to emissions from pollutant $E$ by the emission change ratio $E_{2002}/E_{2010}$ on a country basis.

As for the Appendix, deposition data have been extrapolated to the equivalent of 100% emission reductions. See comments above and in section 4.6.2, for the validity of this procedure.

### 4.11 Summary and conclusions

This chapter has summarised some of the many tests conducted over the last year, designed to assess the behaviour of source-receptor (SR) relationships calculated with the EMEP model. Following the conclusions of the NONLIN project, it has been decided that for this years results SR relationships require four runs of the EMEP model, for emissions of S, N+P, A and V (SO$_x$, NO$_x$, primary particulates, Ammonia and VOC, respectively), for each country or region under analysis.

Tests have been carried out both for depositions and for air concentrations to determine whether the sum of responses to emission changes from individual countries adds up to the response to emission changes when all country emissions are changed at the same time. The overall result of these tests is that the consequences of an emission
change around 15% by several countries can be obtained by simply adding the changes caused by the individual countries. At this level the nonlinear effects can be neglected.

In order to test the applicability of the linear equation for large emission changes we have compared the results obtained using equation 4.1 with the results obtained by direct calculation (i.e including all nonlinear effects). Even if there are non-negligible differences between the two sets of results, the conclusion from these tests is that, especially for depositions, the 2010 CLE results can be extrapolated to relatively large emission changes with some confidence.

We have also investigated the way in which \( \frac{\partial X}{\partial E} \) and SR matrices change depending on the base-case or reference emission regime from which they are calculated. A general finding is that model derivatives obtained at CLE emission levels may need to be modified for other emission regimes, especially for ozone and PM components. The main modifying factors are associated with the NO\(_x\) and Ammonia levels. Ozone and its derived indicators are most sensitive to NO\(_x\) and VOC emissions, as already established from previous studies. Concentrations of fine particulate nitrate and ammonium are sensitive to three precursors, SO\(_x\), NO\(_x\) and Ammonia, with only VOC emissions having little effect.

These results and many others need to be examined in more detail before specific recommendations can be made concerning applications of such derivatives in integrated assessment models, but they also suggest that in most cases the changes in derivatives are relatively modest.

The results presented here justify the choice of source-receptor calculations as presented in the Appendix.
CHAPTER 4. SOURCE-RECEPTOR CALCULATIONS

References


Air concentrations and depositions of acidifying and eutrophying components, status 2002

Hilde Fagerli

In this chapter we present model results for 2002 and compare them to measurements in the EMEP network. Moreover, we show modelled depositions and use the depositions to different ecosystems to calculate exceedances of critical loads in Europe. Finally, we discuss how the model results for 2002 presented here differ from results presented in earlier EMEP reports.

In EMEP Status report I, part II from last year (2003) we presented an extensive evaluation of the acidifying and eutrophying components for the years 1980, 1985, 1990 and 1995 to 2000. In EMEP Status report I, part III (2003) we showed a comparison of observations and modelled results for 2001. In this report we present main results for 2002 with the most updated EMEP Unified model, version 2.0. This version differs slightly from last years version, as described in chapter 2. The changes in the model have resulted in a lowering of the aerosol nitrate concentrations in air and model results in better agreement with the observations. However, the results from revision 1.8 and 2.0 are otherwise very similar. Thus, the old evaluation of the model is still valid. Therefore, only a brief analysis of the model performance for acidifying and eutrophying components is given here, highlighting the differences and similarities to previous results.
5.1 Comparison of observations and model results for 2002

In order to obtain data that are characteristic for long-range transport, measurements in the EMEP network are collected at background stations that is representative of a larger area. The size of this area is determined by the variability of the air and precipitation quality and the desired spatial resolution in the concentrations and deposition fields. For the particular purpose of the model validation, measurements should also be representative of the EMEP 50×50 km² grid square average. However, the recommendation for the site not to be influenced by local pollution implies that their location is chosen to ensure representativeness of the minimum concentration in the grid, not the grid average.

The agreement between model predicted and observed air and precipitation data also depend upon an adequate description of emissions. This includes both a reasonable estimates of national totals, sector data and gridded sector data distribution as well as the temporal distribution of emissions. From the reasoning above it is clear that the agreement between model results and observations depend on a combination of several elements; the quality and representativeness of the measurement sites, the adequacy of emissions and the model performance. Therefore, the following discussion on model underestimation and overestimation simply imply that the calculated values are lower or higher than the observations and does not refer to model deficiency only.

5.1.1 Sulphur dioxide in air

In figure 5.1 we show scatter-plots for yearly averaged values for sulphur dioxide concentrations in air. The correlation between the yearly averaged observed and modelled data at EMEP stations is high (correlation coefficient of 0.67), showing that the model captures reasonably well the spatial distribution of the high and low concentration areas – though this is mainly a reflection of emissions distribution. In general, the model overestimates the SO₂ concentrations, in average by 33 %.

In figure 5.2 a map of modelled sulphur dioxide concentrations in air for 2002 is compared to an SO₂ field kriged from measurements (A short description of the kriging method can be found in Hjellbrekke et al. (1997)). The model does to a large extent reproduce the spatial pattern with decreasing concentrations toward the north, high concentrations at the north coast of Spain and in east Europe. However, in general it overestimates the sulphur dioxide concentrations somewhat.

In figure 5.3 frequency analysis for modelled versus observed daily values of SO₂ concentrations is shown. The model shows a tendency to give too many days with high concentrations and too few days with low concentrations compared to the measurements. This is consistent with the conclusions from last years report that summer concentrations of SO₂ are captured very well by the model, whereas the higher winter concentrations are overestimated. This can also be seen from monthly time-series of
CHAPTER 5. SULPHUR AND NITROGEN

Figure 5.1: Scatter-plots of modelled versus observed concentrations in air of sulphur dioxide, sulphate, ammonia and sum of ammonia and ammonium (in $\mu g(S) m^{-3}$ and $\mu g(N) m^{-3}$)

model results versus observations shown in figure 5.4. Here we show results which are averaged over all the stations which have reported daily data for 2002, but the same conclusions are valid for the majority of stations if studied separately.

5.1.2 Sulphate in air

Scatter-plot for modelled versus measured sulphate concentrations in air for 2002 is presented in figure 5.1. 99% of the annual mean concentrations for the different sites
Figure 5.2: Comparison of model results and krieded observations for SO$_2$ in air ($\mu$g(S) m$^{-3}$), 2002
Figure 5.3: Frequency analysis of modelled versus observed concentrations in air of sulphur dioxide, sulphate, ammonia and sum of ammonia and ammonium (in $\mu g(S) \ m^{-3}$ and $\mu g(N) \ m^{-3}$)

are within a relative bias of 50% and approximately 96% within relative bias of 30%. The spatial correlation coefficient is high, 0.78. The daily correlations between model and measurements (not shown) are among the highest for the acidifying and eutrophying compounds. This is partly due to sulphate being a secondary pollutant and thus less dependent on the ability of the model to simulate dispersion close to the sources. In addition the data quality of measurements of concentrations of $SO_4^{2-}$ in air is among the best. In figure 5.3 frequency analysis of daily sulphate concentrations are compared.