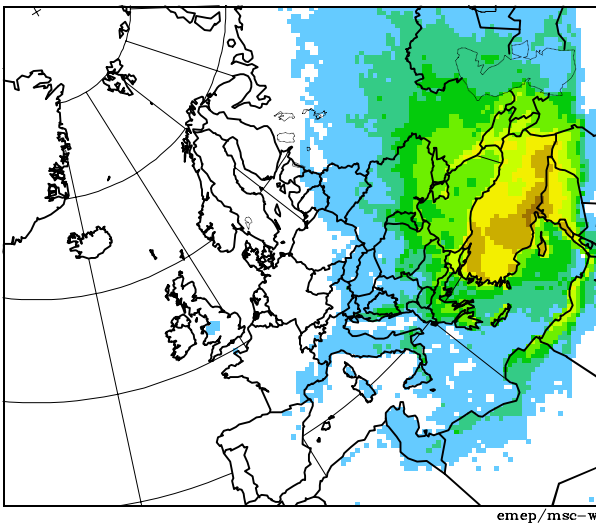


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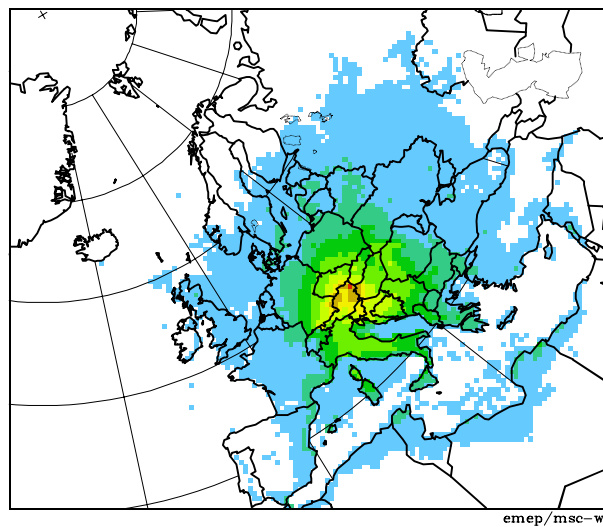
Co-operative programme for monitoring  
and evaluation of the long range  
transmission of air pollutants in Europe

## Transboundary Acid Deposition in Europe

1997 deposition of oxidized nitrogen from Turkey



1997 deposition of oxidized nitrogen from Austria



**EMEP SUMMARY REPORT 1999**

Edited by L. Tarrasón and J. Schaug

# CCC & msc-w

EMEP

Report 1/99

Date:

July 1999

DET NORSKE METEOROLOGISKE INSTITUTT  
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Research Report no. 83

# **TRANSBOUNDARY ACID DEPOSITION IN EUROPE**

EMEP SUMMARY REPORT  
1999

Edited by  
Leonor Tarrasón and Jan Schaug

ISSN 0332-9879



# Transboundary Acid Deposition in Europe

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## *Preface*

The Convention on Long Range Transboundary Air Pollution (LRTAP) provides a framework for international action to reduce the impact of air pollution. The work under the Convention has established a sound a process for negotiating concrete measures to control emissions of air pollutants through legally binding protocols. In this process, the main objective of the EMEP programme (Co-operative Programme for Monitoring and Evaluation of the Long-Range Transmission of Air pollutants in Europe) is to regularly provide with qualified scientific information to support the review and further extension of the international protocols negotiated within the Convention.

The main task of EMEP has been to provide the ECE Governments and the Convention on Long-range Transboundary Air Pollution with regular information on past and predicted emissions, concentrations and/or depositions of air pollutants in the EMEP region and, in particular, on the quantity and significance of their long-range transboundary transport. As the work of the Convention has advanced, the requirements on the information provided by EMEP have also evolved. On the seventh phase of the programme, EMEP has been required to focus further in the evaluation of international abatement strategies and review the successes or failures of the existing Protocols. To satisfy these needs, the work of EMEP under the Convention has been formulated in five thematic areas (ref. EB.AIR/GE.1/1998.3):

- Acid deposition and eutrophication
- Photochemical oxidants
- Heavy metals
- Persistent organic pollutants
- Aerosols

This report focuses on the first of these areas: “Acid deposition and eutrophication” and has been prepared for presentation at the twenty third session of the Steering Body of EMEP.

It summarises the 1997 European situation with respect to acidifying and eutrophying pollution transport across national boundaries and qualifies the assessment by reviewing the current achievements and limitations of the EMEP data. The analysis is intended to fulfil the main objectives for the acidification and eutrophication programme. Therefore, this report:

- a) presents a survey of the progress towards the reduction of emissions and depositions, in particular as it concerns exceedances of critical loads,
- b) quantifies transboundary fluxes allocating the sources of the estimated depositions,

- c) provides sulphur and nitrogen deposition values in a form useful for the assessment of effects, and
- d) reports progress in the development of tools to provide guidance on the most cost-effective way of attaining environmental goals.

Conclusions are presented both for the whole EMEP area and for each individual Party to the Convention.

There are 44 Parties to the Convention to this date: 41 European countries, Canada, the United States and the European Community. Summary reports are presented in the Appendix, for all Parties except for Georgia and Malta. Georgia was not included in this year's calculations because unfortunately the information that the country had ratified the CLRTAP on February 2nd 1999 did not reach MSC-W in time for the model calculations. The reason not to include Malta is that there is no information available from the country. Malta has not reported emissions, does not participate in the EMEP monitoring programme and the estimated emissions from Malta are so low that its deposition is below the precision limits of the model calculation of transboundary fluxes.

We have made an effort to present this information in a form useful for the Executive Body of the Convention on Long-Range Transboundary Air Pollution, its subsidiary bodies, and the Parties to Convention. It is anticipated that information on the relative importance of national and external emissions will help national authorities estimate their local and regional targets. It is also intended that EMEP results will be useful for the Working Group of Effects and the Working Group of Strategies and Review and thus form a basis for further evaluating international abatement strategies.

The work of EMEP is carried out in collaboration with a broad network of scientists at national level that contribute with the systematic collection, analysis and reporting of emission inventories, measurements from the EMEP monitoring networks and results from modelling studies. These contributions are essential to EMEP and the success of the programme relies on this participation. The present report is intended to stimulate communication between the participants and users of EMEP, and facilitate the exchange of information by clarifying the operational objectives of the programme.

This report consists of three parts. The first part (chapter 1) reviews the European situation with respect to acidifying and eutrophying air pollution since 1985 and presents updated 1997 emissions, depositions, exceedances of critical loads and transboundary source-receptor matrices for the whole EMEP area.

The second part (chapters 2-4) evaluates the requirements of the data used in the EMEP program for acidification and provides suggestions for the future development of the program. Chapter 2 presents an overview of the requirements on EMEP emission data and discusses the completeness of the reported data in relation to these requirements. Chapter 3 evaluates the present status of EMEP monitoring network for acidification and eutrophication and gives recommendations for improvement. Chapter 4 reviews the progress with the EMEP atmospheric dispersion models in terms of their performance and present recommendations for future model development. All three chapters have a similar structure, intended to clarify the requirements on data quality from EMEP and evaluate the 1997 status of the information compiled by the programme.

The third part (appendix) presents the information compiled by EMEP in the form of a review organised by country. For each Party to the Convention, a detailed review of the evolution of the emissions, depositions and critical load exceedances is presented, together with the analysis of its transboundary pollution exchange. Specific comments and recommendations are given as necessary, on the status of the national EMEP monitoring network and of the reported emissions.

The overall conclusion is that the EMEP programme has an impressive compilation of qualified data on transboundary acidifying pollution in Europe and, despite its limitations, is well in position to achieve its goals for the seventh phase.

All data included in this report will be available via Internet after it is approved by the Steering Body of EMEP. Countries are encouraged to analyse the data and provide their own conclusions. Reactions and comments are both welcome and encouraged.

<http://www.emep.int>



***Transboundary Acid Deposition  
in Europe in 1997***



## ***1. Transboundary acid deposition in the EMEP area***

The international commitment to reducing acidification was the driving force behind the adoption of the Convention on Long Range Transboundary Air Pollution in 1979. At present, a substantial decrease on acid deposition has been recorded throughout Europe and both the actual exceedances and the area of ecosystems exposed to exceedance of critical load have been considerably reduced. Eutrophication, on the other hand, still remains a problem in most of the EMEP area.

This chapter gives an overview of the 1997 status of transboundary acidifying and eutrophying pollution in Europe and traces the progress towards environmental improvements, measured in terms of exceedances to critical loads. To that purpose, trends in emissions, depositions, transboundary depositions and critical load exceedances since 1985 are presented and discussed. The European scale analysis performed here is complemented with a detailed analysis per country that is presented in the Appendix.

Special attention is given to the quantification of transboundary depositions, which for the first time, are calculated with the 20-layer EMEP Eulerian Acid Deposition model. Updated 1997 geographical distributions of depositions and conditional exceedances to critical loads are presented in the extended 50x50 km<sup>2</sup> EMEP grid.

Measurements are not directly included in the present assessment, although it is intended to do so in future analysis. The study of trends in EMEP monitoring data demands a considerable quality control effort which has been initiated recently and will be reported in due time.

### ***1.1. Trends of emissions, depositions and critical load exceedances***

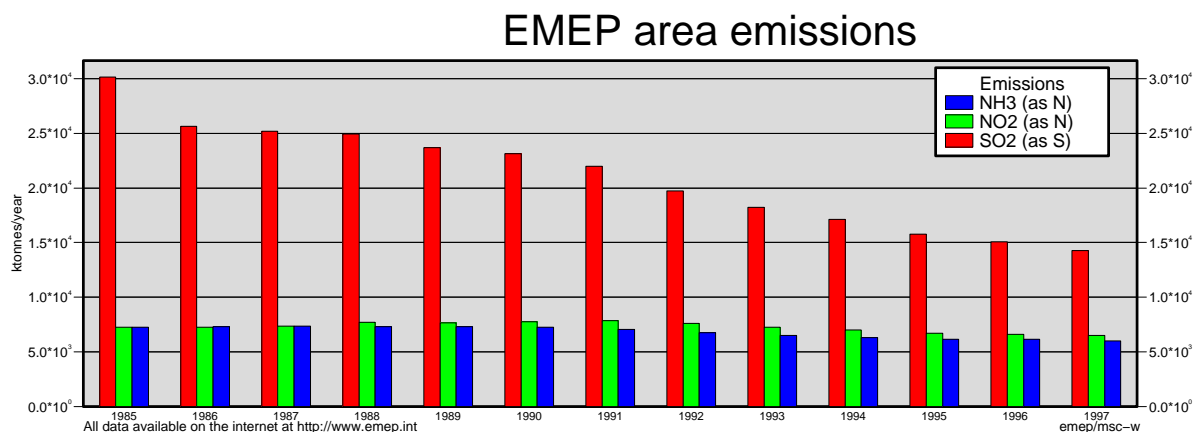
Since 1980, anthropogenic emissions of sulphur dioxide have been reduced in the whole EMEP area by 55%. According to MSC-W estimates, sulphur emissions in the EMEP area had decreased by 30% compared to 1980 levels already in 1989, thus meeting the requirements of the First Sulphur Protocol long before due in 1993. However, the emission reduction for the whole EMEP area is not representative for individual countries and, in 1993, officially reported emission data shows that 9 different countries did not meet the agreements from the First Sulphur Protocol (Mylona, 1999).

The trend in the emission of NO<sub>x</sub> is characterised by relatively high releases in the late 1980s and a steady slow decrease during the 1990s. The 13% reduction in 1997 compared to the 1980 level reaches 17% when 1987 (base year of the NO<sub>x</sub> Protocol) is used as a reference instead.

Emission reductions estimated for the whole EMEP area satisfied in 1994 and 1996 the requirements from the First NO<sub>x</sub> Protocol, but according to the official reported values, 6 countries had larger still NO<sub>x</sub> emissions in 1996 than in 1987.

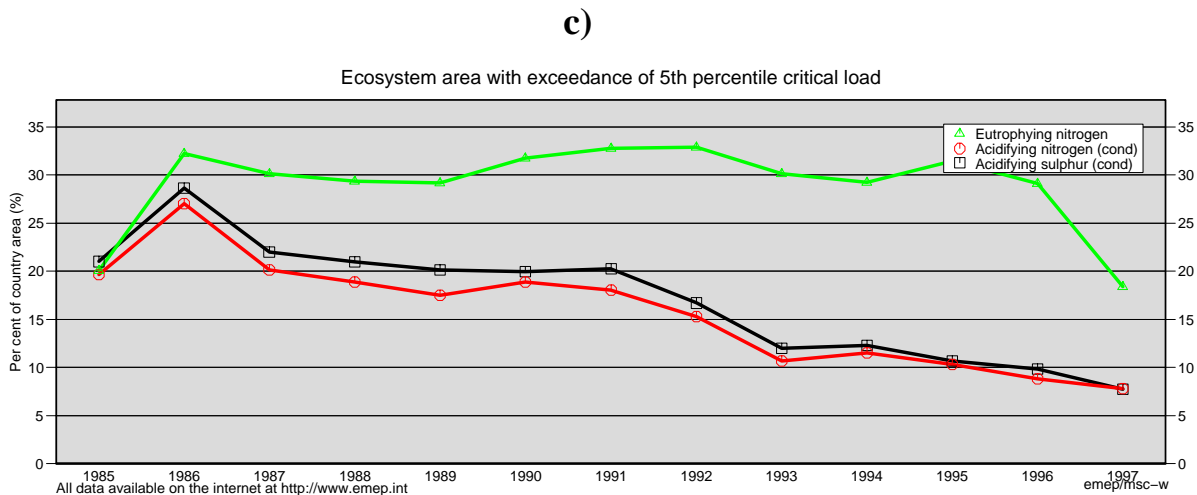
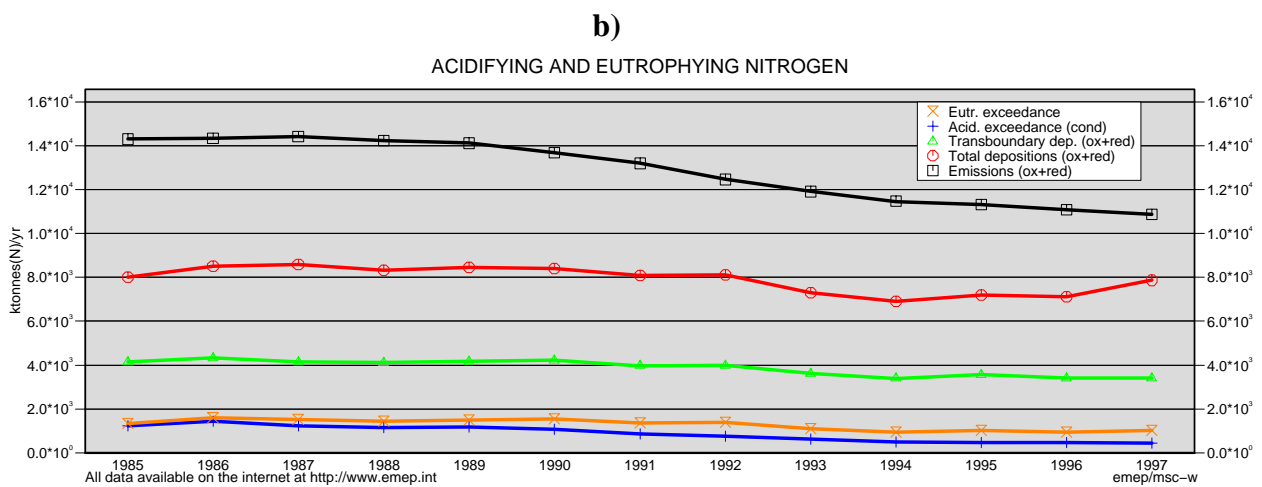
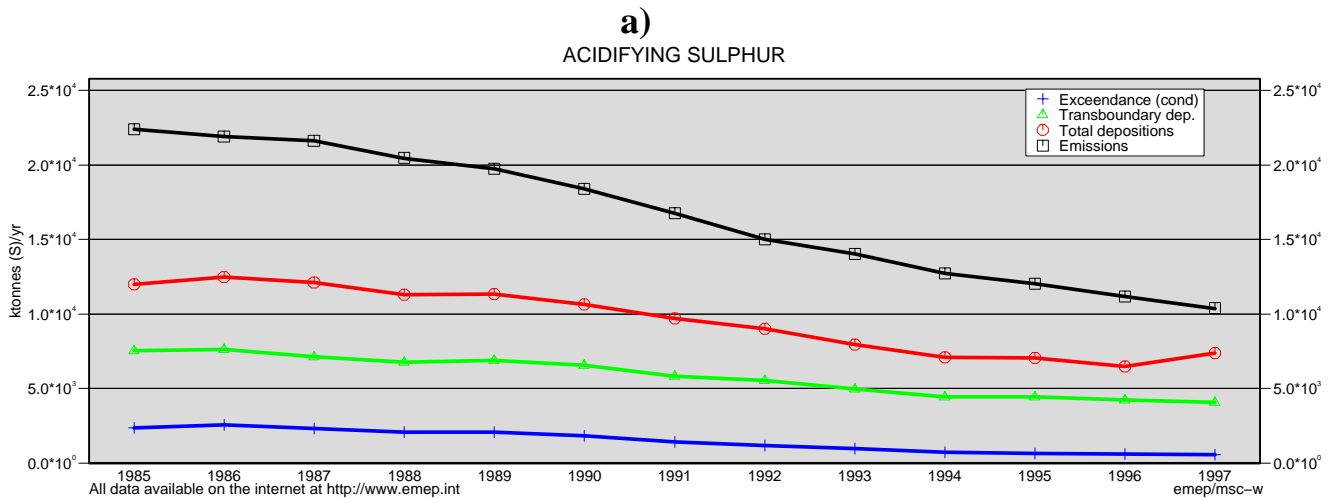
European emissions of ammonia appear to have dropped by approximately 15% between 1990 and 1997. The almost constant emission trend before 1990 is primarily a result of the assumptions made to estimate the large amount of missing data for most countries.

Emission trends have been derived at MSC-W from officially reported emissions and expert estimates. It is not possible at present to provide official emission trends because not all Parties to the Convention have reported national totals for all years since 1980. For SO<sub>2</sub>, only 43% of the countries have reported emission data for all years, and this percentage decreases to 41% for NO<sub>x</sub> and 16% for ammonia. National emission trend values are given in Tables 1.1 -1.3 at the end of this chapter and visualised in Figure 1.1. for trends since 1985. These are the values that have been used for the assessment summarised in the following.



**Figure 1.1.** Estimated emission trends since 1985 for the whole EMEP area used in acidification model calculations (emission values by country are given in Tables 1.1-1.3; adapted from Mylona, 1999).

Trends on emissions, depositions, transboundary depositions, and exceedances to critical loads have been estimated for each Party to the Convention on Long Range Transboundary Air Pollution. The analysis has been performed separately for sulphur and nitrogen compounds, using conditional critical loads calculated by the Coordination Centre for Effects (CCE). In addition, the ecosystem area in each country exposed to exceedances of 5th percentile critical load has been calculated as percentage of the total area of the country. This information is given in the Appendix, using three different figures for each country.



**Figure 1.2. a)** Trends of total sulphur deposition (red), transboundary deposition (green) and acidifying conditional exceedance (blue) for countries in EMEP area.

**b)** trends of total nitrogen deposition (red), transboundary deposition (green), acidifying conditional exceedance (blue) and eutrophying exceedance (yellow) for countries in EMEP area. Sulphur and nitrogen emissions (both as  $\text{NO}_x$  and  $\text{NH}_3$ ) are given in black curve.

Units: 1000 tonnes (S or N)/year.

**c)** Ecosystem area with exceedance of critical load as percentage of the total country area.

The first figure, for acidifying sulphur, shows the total sulphur mass deposited over the country each year and shows the effect of this deposition as the total mass contributing to exceedances of the 5th percentile conditional critical load. In general, trends in the exceedances follow the trends in sulphur and nitrogen deposition.

The acidifying sulphur figure includes also a curve depicting the time evolution of deposition of transboundary origin to the specific country. Differences between total deposition and transboundary deposition give a good measure of the importance of indigenous deposition in the country. In many cases, specially over central European countries, it can be seen how the relative contribution of transboundary deposition has increased after the emissions from the countries themselves have decreased. Direct comparison between the emissions and depositions values allows a first analysis of the country balance of imported vs exported deposition.

The second figure serves for the analysis of acidifying and eutrophying nitrogen. Excess nitrogen deposition contributes to acidification but can also lead to the eutrophication of soils and surface waters. Thus, to analyse the effects caused by excess nitrogen deposition both a conditional critical load for acidity and a critical load of nutrient nitrogen need to be considered.

The conditional critical load for acidifying nitrogen depends both on sulphur and nitrogen deposition, and thus the acidifying nitrogen exceedance trend depends both on sulphur and nitrogen deposition trends. The critical load for eutrophying nitrogen does not depend on sulphur deposition and therefore the eutrophying exceedances follow more generally the nitrogen depositions.

The third figure gives the time evolution of the area of ecosystems with exceedance of the 5th percentile critical loads. The curves represent the time evolution of the ecosystem area exposed to possible exceedances, as percentage of the total country area. Here, the geographical distribution of the considered ecosystems plays an important role. Inter-annual meteorological variations can affect the pollution transport patterns, resulting in large changes in the percentage ecosystem area with exceedances of the 5th percentile. Larger deposition amounts over the country, do not necessarily imply larger ecosystem area with registered exceedances because both the deposition levels and the geographical distribution of sensitive ecosystems determine the changes in the ecosystem area.

Figures 1.2 a), b) and c) summarise the analysis for all countries in Europe that are Parties to the Convention except Georgia, Iceland, Liechtenstein, Malta and Turkey. Georgia was not included in this year's calculations because the information that the country had ratified the CLRTAP on February 2nd 1999 did not reach MSC-W in time for the model calculations. The emissions from Malta and Liechtenstein are so low that their depositions are below the precision limits of the model calculation of transboundary fluxes. Finally, Iceland and Turkey are not included either because there is no critical load data available for these countries.

The transboundary deposition in these figures is the sum of total deposition in the individual countries that originates outside of their boundaries. For all these EMEP countries, transboundary depositions represent between 55-60% of the total deposition for sulphur, and between 40-50% of the total deposition for nitrogen. Although emissions have decreased, the distribution of sources in the different countries has not given the same type of reduction of in the depositions.

Since 1985, exceedances to conditional critical loads have decreased by 76% for sulphur and by 65% for acidifying nitrogen. The decrease is mostly due to the reduction of sulphur emissions and depositions, because nitrogen depositions have been kept almost constant during the period. Consequently, there is no significant change in the total eutrophying exceedances for these areas.

The percentage ecosystem area with exceedance of 5th percentile critical loads for acidification has steadily decreased, from 21% of total country area in 1985 to 8% in 1997. In contrast, the ecosystem area exposed to eutrophication has been kept almost constant during the period. Note here that although nitrogen deposition increased in 1997 the percentage ecosystem area with exceedance in fact decreased this year, because of the particular geographical distribution of the depositions and the sensitive ecosystems.

## ***1.2. Geographical distribution of acidifying and eutrophying deposition and exceedances***

The trends presented in the previous section have been calculated with the EMEP Lagrangian model up to 1996. 1997 values have been calculated with the EMEP Eulerian Acid Deposition model which is operative for EMEP for the first time this year. The scientific challenges associated to the introduction of the operational Eulerian EMEP Eulerian Acid Deposition are discussed in detailed in Bartnicki (1999) and Olendrzynski (1999). A short review of these is also given in Chapter 4.

The main advantages for the work of the Convention derived from the introduction of the operative Eulerian model are:

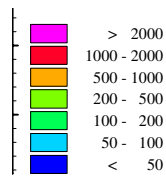
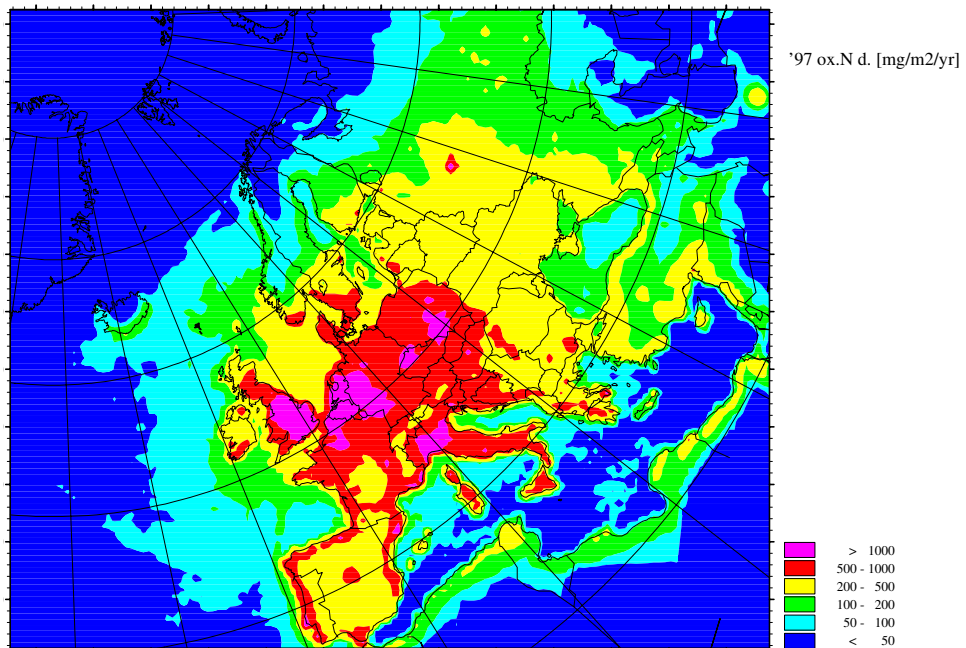
- \* the increased spatial resolution of the new Eulerian model , which has been verified to improve the level of accuracy of the EMEP estimates,
- \* the improved consistency in the allocation of sources and calculation of transboundary fluxes
- \* the fact that it represents a first step towards the implementation of a new generation of integrated multi-component models for policy applications

An additional improvement is the extension of the EMEP grid in order to fully include in the calculations new Parties to the Convention, like Cyprus, Turkey, Armenia and Georgia, and areas of general interest like the Mediterranean Sea.

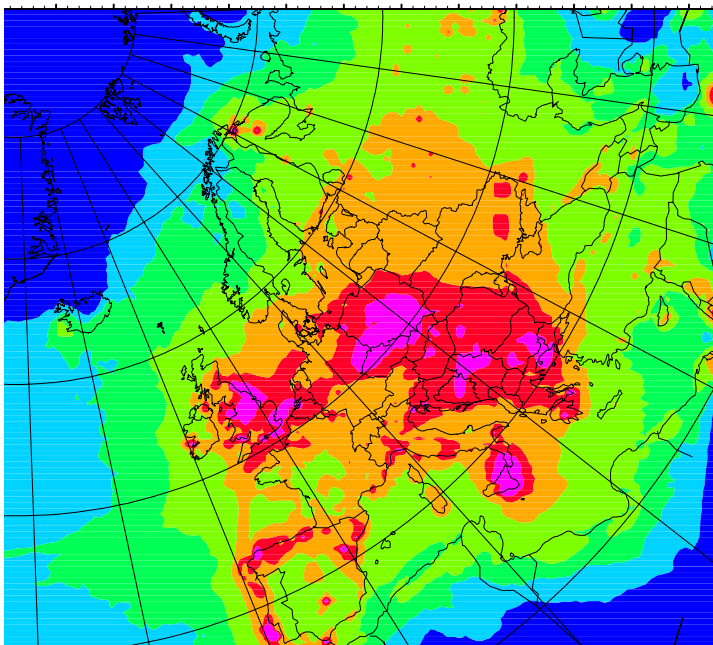
It is expected that the increased accuracy and improved spatial resolution from the EMEP Eulerian Acid Deposition Model will be useful for scientific research in the Working Group of Effects, for integrated assessment modellers, and for researchers and authorities at national level.

Figures 1.3. a), b) and c) show the 1997 total deposition fields for the major acidifying and eutrophying compounds. Overall, an increased level of detail due to better spatial resolution and multilayer structure is attained.

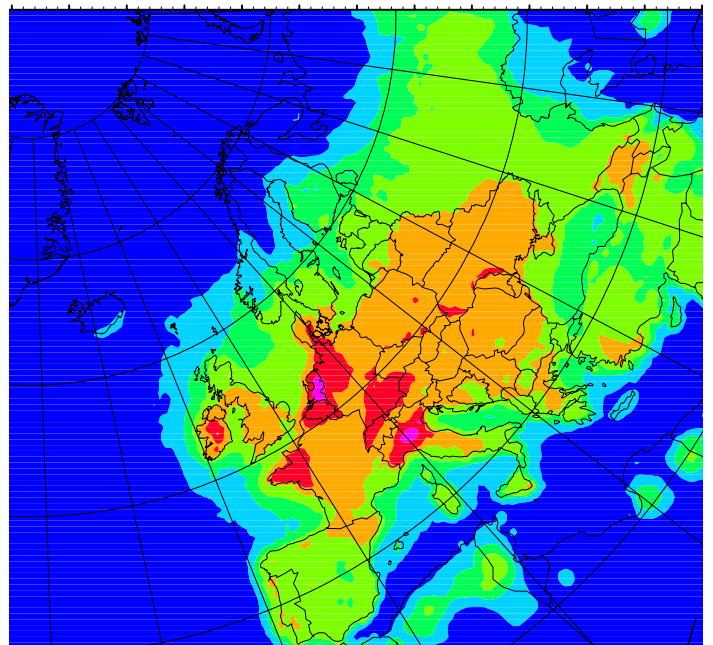
a)



b)



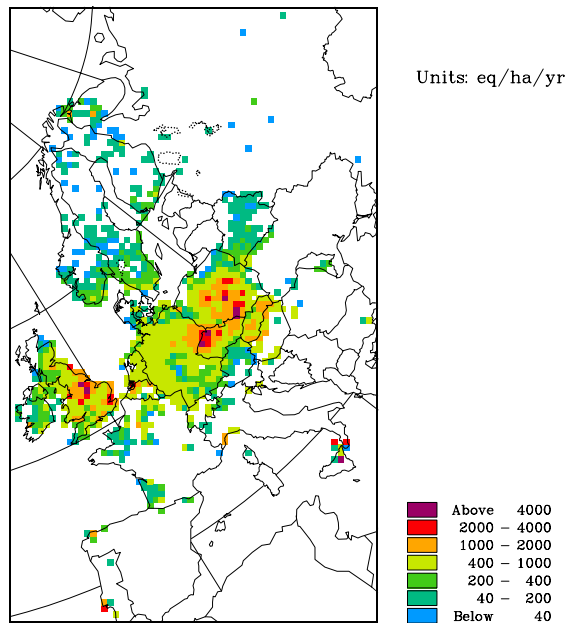
c)



**Figure 1.3** a) 1997 total deposition of oxidised nitrogen ( $\text{mg(N)/m}^2/\text{a}$ )  
b) 1997 total deposition of oxidised sulphur ( $\text{mg(S)/m}^2/\text{a}$ )  
c) 1997 total deposition of reduced nitrogen ( $\text{mg(N)/m}^2/\text{a}$ )  
\* Scale for b) and c) given in the centre of the page

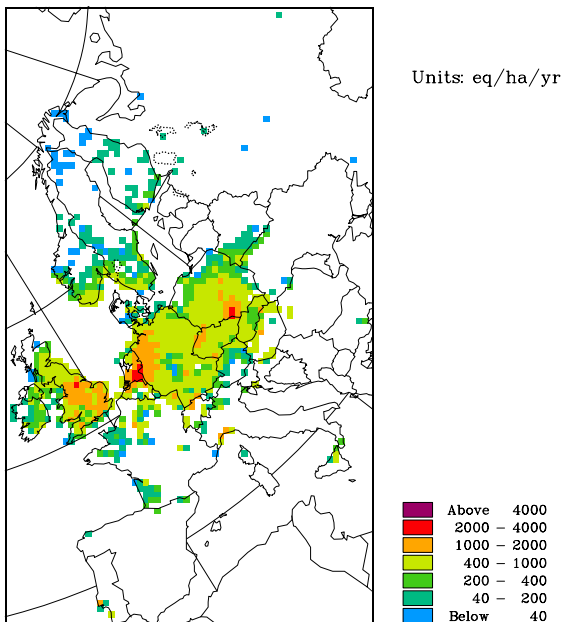
a)

Exceedances of the 5 percentile CL(S|Ndep97)  
EMEP Eulerian model. Critical loads from CCE



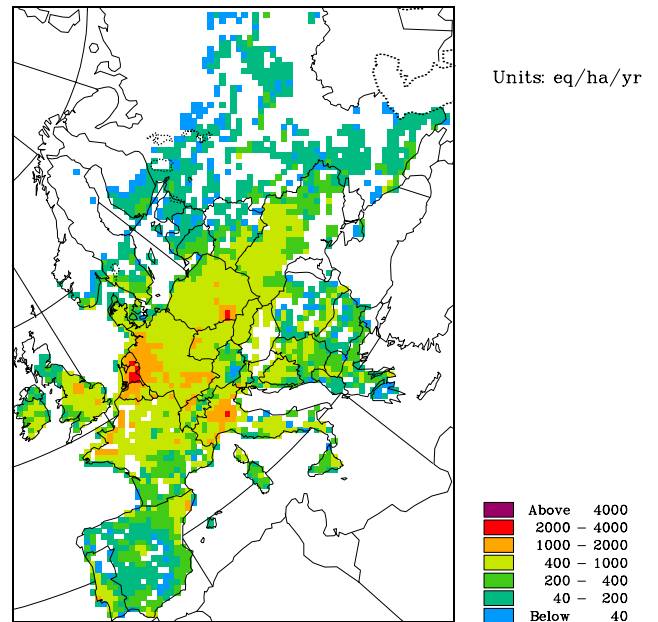
b)

Exceedances of the 5 percentile CL(N|Sdep97)  
EMEP Eulerian model. Critical loads from CCE



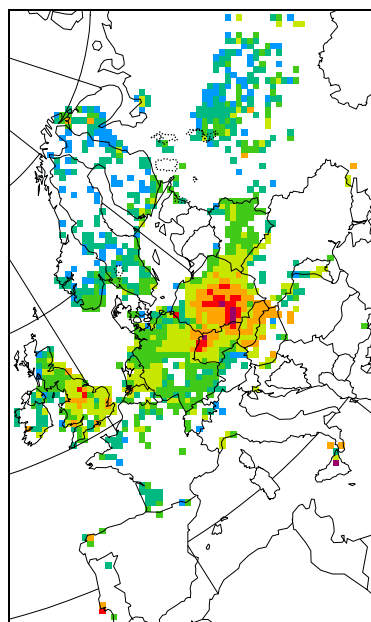
c)

Exceedances of the 5 percentile CL(Nut) 1997  
EMEP Eulerian model. Critical loads from CCE

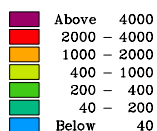


**Figure 1.4** 1997 exceedances of the 5th percentile critical loads for:  
a) acidifying sulphur (conditional) based on CL(S|Ndep97)  
b) acidifying nitrogen (conditional) based on CL(N|Sdep97)  
c) eutrophying nitrogen based on CL(Nut)  
Critical Loads calculated by CCE

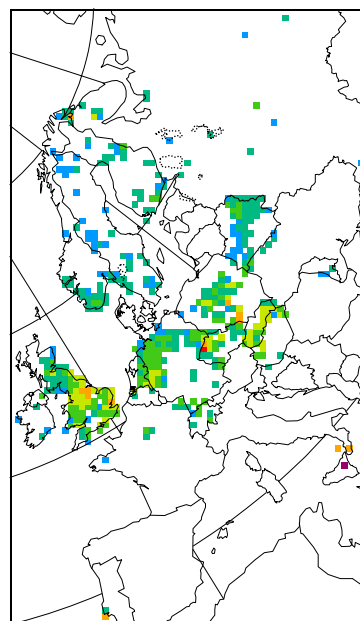
Exceedances of the 5 percentile CL(S)Ndep10off)  
EMEP Eulerian model. Critical loads from CCE



Units: eq/ha/yr



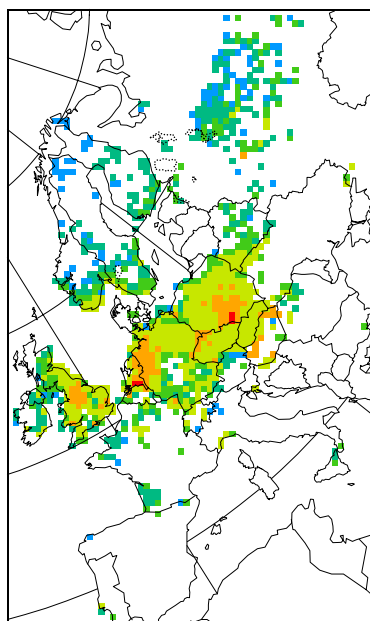
Exceedances of the 5 percentile CL(S)Ndep10G52)  
EMEP Eulerian model. Critical loads from CCE



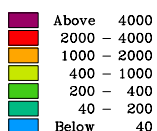
Units: eq/ha/yr



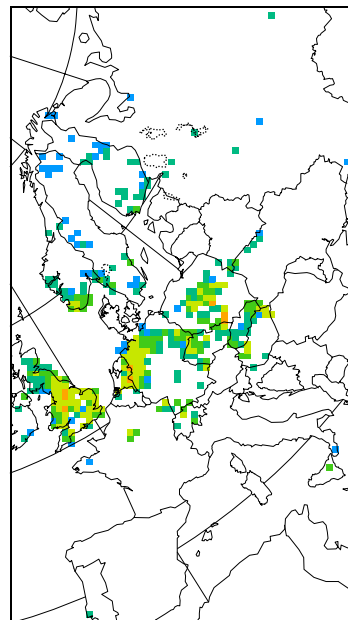
Exceedances of the 5 percentile CL(N)Sdep10off)  
EMEP Eulerian model. Critical loads from CCE



Units: eq/ha/yr



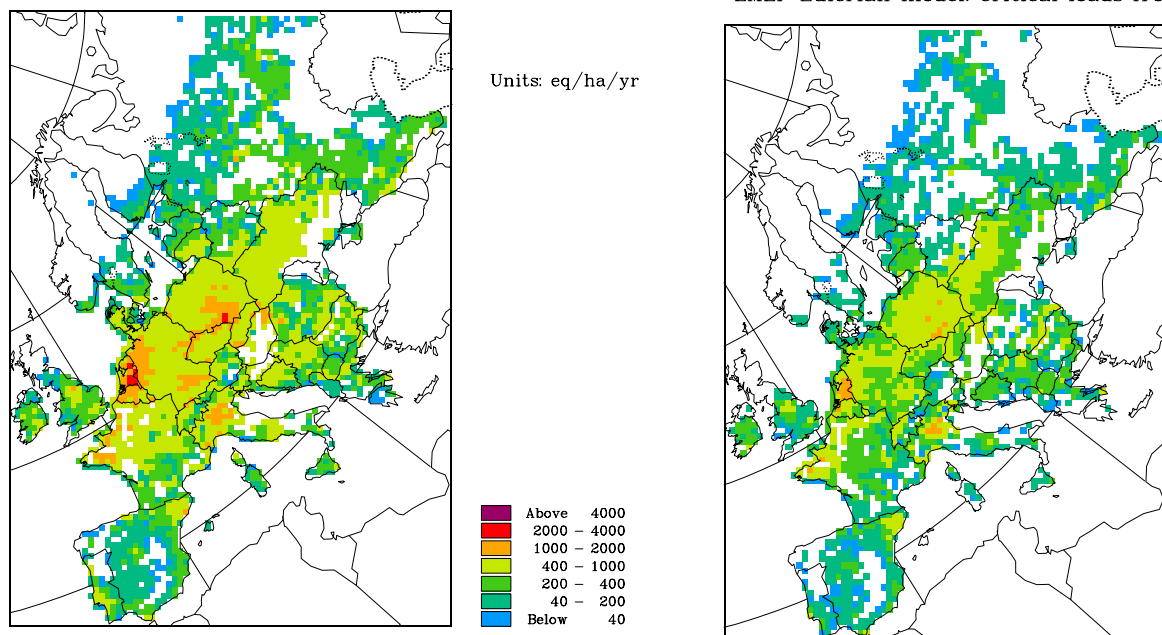
Exceedances of the 5 percentile CL(N)Sdep10G52)  
EMEP Eulerian model. Critical loads from CCE



Units: eq/ha/yr



**Figure 1.5** Comparison of the 5th percentile exceedances for acidifying sulphur (above) and acidifying nitrogen (below) for two different emission projection scenarios. Left figures are based on the official reported 2010 emission projections. Right figures are based on scenario G5/2 for integrated assessment. Critical loads calculated by CCE.



**Figure 1.6** Comparison of the 5th percentile exceedances for eutrophying nitrogen (below) for two different emission projection scenarios. Left figure is based on the official reported 2010 emission projections. Right figure is based on scenario G5/2 for integrated assessment. Critical loads calculated by CCE.

The geographical distribution of maximum depositions and their absolute values are comparable to those from previous estimates by the Lagrangian model. Differences between the models are within the expected meteorological variability. Largest differences are found for the deposition of nitrogen oxides (figure 1.3.a) where the Eulerian model estimates larger dry deposition values close to source areas.

The increased spatial resolution in model calculations allows for a calculation of exceedances to critical loads in the  $50 \times 50 \text{ km}^2$  grid (Posch et al., 1999). As expected, the spatial distribution of exceedances changes with respect to previous estimates with the Lagrangian model. Both geographical position of maximum and the value of the maximum exceedance are similar in the two models. There is no systematic pattern as the exceedances depend on the geographical distribution of sensitive ecosystems as well as on the actual deposition of sulphur and nitrogen. Note for example the increase of exceedances in Southern European countries, which is due in part to the increase of the deposition calculated by the Eulerian model in these areas in 1997.

Figures 1.5 and 1.6 compare the 5th percentile exceedances of conditional critical loads for acidification and for eutrophication derived from two different emission projection scenarios. It is interesting to note that the exceedances calculated with the official 2010 emission projection scenario are considerably higher than the exceedances derived from the G5/2 scenario. In fact, the 2010 official projection exceedances are larger around the maxima in the Black Triangle than the 1997 estimates. This behaviour is a consequence of the poor quality of

emission projection data. Below 40% of the Parties to the Convention have reported emission projections for the three requested years and the reporting of these projections is not satisfactorily harmonised. In this situation is rather important for EMEP to co-operate with integrated assessment modellers that can assist in the calculation of the emission projections.

### 1.3. Transboundary fluxes for 1997

The computation of transboundary fluxes for 1997 has been carried out for the first time with the Eulerian Acid Deposition model, for the different emitting regions described in Table 1.4

**Table 1.4 :** Emitting regions displayed, and their identifying codes.

Country/Region	Code	Country/Region	Code
Armenia	AM	Norway	NO
Austria	AT	Poland	PL
Belarus	BY	Portugal	PT
Belgium	BE	Republic of Moldova	MD
Bosnia and Herzegovina	BA	Romania	RO
Bulgaria	BG	Russian Federation	RO
Croatia	HR	Slovakia	SK
Cyprus	CY	Slovenia	SI
Czech Republic	CZ	Spain	ES
Denmark	DE	Sweden	SE
Finland	FI	Switzerland	CH
France	FR	The FYR of Macedonia	MK
Georgia	GE	Turkey	TR
Germany	DE	Ukraine	UA
Greece	GR	Yugoslavia	YU
Hungary	HU	United Kingdom	GB
Iceland	IS	Remaining Land Areas	REM
Ireland	IE	The Baltic Sea	BAS
Italy	IT	The Mediterranean Sea	MED
Latvia	LV	The North Sea	NOS
Lithuania	LT	Remaining N.E. Atlantic	ATL
Luxembourg	LU	Natural Oceanic	NAT
Malta	MT		
Netherlands	NL	European Union	EU

In Table 1.4 Russian Federation means the part the Russian Federation inside the EMEP domain of calculations. The same applies to the Remaining N.E. Atlantic region and Natural Oceanic emission area. Remaining Land Areas include North Africa, Albania, Estonia, Kazakhstan, Azerbaijan, Syria, Lebanon, Israel, parts of Uzbekistan, Turkmenistan, Iran, Iraq and Jordan. European Union includes 15 countries already listed in Table 1.4: Austria, Belgium, Denmark, Finland, France, Germany, Greece, Ireland, Italy, Luxembourg, The Netherlands, Portugal, Spain, Sweden and United Kingdom.

The accuracy of the source-receptor matrices and the transboundary fluxes calculated by the Eulerian model is discussed in Bartnicki (1999). In general, the Eulerian model allocates a larger fraction of the European emissions. This is well illustrated in Table 1.5 where the 1997 import-export budgets for each country/region is summarised. Averaged over the EMEP area, the Eulerian model manages to allocate 90% of the emitted mass of oxidised sulphur, 82% of oxidised nitrogen and 95% of the emitted mass of reduced nitrogen. The rest of the emissions are transported out of the EMEP area boundaries.

**Table 1.5: Import-Export Budgets for 1997**

Receptors	Oxidized sulphur						Oxidized Nitrogen						Reduced Nitrogen					
	Export		Import		Sea	EMEP	Export		Import		Sea	EMEP	Export		Import		Sea	EMEP
	Mass	%	Mass	%	%	%	Mass	%	Mass	%	%	%	Mass	%	Mass	%	%	%
AT	226	79	698	92	15	95	447	85	420	85	9	87	380	61	487	67	6	99
BE	1037	86	272	62	33	97	889	87	275	68	24	90	589	74	180	46	24	99
BG	5646	83	970	45	25	87	513	75	294	63	13	89	377	59	340	57	10	89
DK	472	87	280	79	46	98	718	95	258	87	25	78	622	74	131	38	44	100
FI	299	60	802	80	23	98	601	76	343	64	11	69	135	48	165	53	17	99
FR	3595	70	1981	56	34	95	3134	63	1707	48	21	89	2320	42	725	19	22	98
DE	5629	77	2174	56	22	96	3919	71	1627	51	15	87	2591	49	1089	28	14	99
GR	2278	84	1092	71	33	79	933	82	301	59	21	78	532	60	180	34	24	89
HU	2689	82	863	59	15	94	445	74	408	72	6	94	391	63	351	60	4	97
IS	100	82	68	75	76	97	79	91	59	88	40	61	16	66	17	67	60	98
IE	623	76	207	51	59	98	345	92	250	89	47	79	603	55	95	16	40	99
IT	4967	75	2253	58	36	90	3792	70	693	30	17	85	1772	47	288	12	19	96
LU	38	95	28	93	25	97	63	94	27	88	17	89	49	85	27	76	12	99
NL	512	83	320	75	38	98	1273	89	269	63	27	86	814	68	240	39	24	99
NO	103	68	715	94	42	99	581	86	430	82	30	69	117	54	211	68	31	99
PL	5993	55	2353	32	11	98	2291	65	1248	50	8	87	1324	46	953	38	7	99
PT	1529	82	217	39	48	80	958	77	221	44	23	68	499	63	74	20	26	83
RO	3108	68	2072	59	16	93	714	74	654	72	9	86	902	50	704	43	8	97
ES	7469	72	595	17	35	86	2254	61	682	32	19	84	1286	45	280	15	18	91
SE	226	65	1113	90	36	99	662	78	806	81	18	75	307	58	387	64	31	99
CH	92	71	270	87	12	96	320	84	177	74	11	87	311	53	181	40	6	99
TR	992	56	2340	75	13	83	1327	63	793	50	8	71	1076	41	637	29	13	90
GB	5655	68	706	21	52	98	4130	73	712	32	35	85	1654	62	352	26	46	99
BY	751	72	1240	81	6	95	486	85	511	85	5	78	860	48	571	38	3	99
UA	3856	68	3416	65	13	92	1042	75	1255	79	8	82	2488	41	1273	27	10	98
MD	77	91	283	97	21	93	84	92	81	92	11	82	280	72	189	64	10	99
RU	4730	39	7957	51	10	84	3817	53	2545	43	4	64	1223	20	2572	35	3	93
LV	252	85	347	89	18	97	100	94	223	97	9	76	100	72	187	82	13	99
LT	317	82	398	85	13	96	161	93	247	95	8	78	186	64	229	69	7	99
CZ	2447	70	1101	51	12	97	1003	78	455	62	8	90	429	64	427	64	7	98
SK	877	87	747	85	13	95	335	89	293	88	6	89	293	71	259	69	4	98
SI	523	87	181	69	18	92	197	92	133	88	11	90	126	71	92	64	10	98
HR	300	75	606	86	25	95	184	81	300	88	11	91	132	67	224	78	12	97
BA	1939	81	505	52	20	91	216	89	278	91	13	91	185	72	194	73	12	91
YU*	1955	75	1148	64	16	92	166	83	402	92	10	93	425	57	319	50	8	95
MK	72	85	287	96	18	87	17	96	81	99	13	80	102	73	88	70	10	93
CY	213	91	15	40	31	74	62	88	18	69	19	74	29	87	8	63	39	81
REM	4746	53	4123	50	9	75	1172	60	2453	76	4	66	2256	38	1120	24	6	83
BAS	603	53	2150	80	55	99	901	84	895	84	24	79	0	0	1014	100	0	100
NOS	983	43	5106	80	70	99	1582	80	2100	84	38	84	0	0	1981	100	0	100
ATL	1945	43	11641	82	60	68	2637	68	3901	76	36	56	0	0	2729	100	0	100
MED	36	60	12538	100	49	79	33	84	2061	100	24	66	0	0	1975	100	0	100
BLS	0	0	2827	100	0	100	0	0	605	100	0	100	0	0	1072	100	0	100
EU	28710	62	6894	28	37	92	18460	55	2931	16	22	84	10456	38	1003	6	23	97

**Export** in tonnes (S or N) and in percent of the country (region) emission.

**Import** in tonnes (S or N) and in percent of the total deposition to the country (region).

**Sea** is the percent of the country (region) emission deposited to the sea surface.

**EMEP** is the percent of the country (region) emission deposited to the EMEP domain.

The following tables show the 1997 source-receptor matrices for oxidised sulphur, oxidised nitrogen and reduced sulphur as calculated by the Eulerian model. Comparison with previous source-receptor matrices calculated with the Lagrangian model show satisfactorily that the Eulerian model manages to account for the “indeterminate deposition” of the previous Lagrangian budget calculations.

The Eulerian model manages to allocate the “indeterminate” contribution thus increasing the actual contribution of different countries. In many cases, the contribution of the country to itself increases in the 1997 simulations. This is particularly true in source areas and for Mediterranean countries. The deposition of Spanish emissions in Spain increases in 1997 with respect to previous years, particularly for oxidised nitrogen and this seems to be related to an increase in the dry deposition calculated by the Eulerian model.

It is not clear at this point whether this is a feature of the Eulerian model or if it is a consequence of the particular meteorological variations during 1997. Still, it can be concluded that differences between the source-receptor matrices produced by Eulerian and the Lagrangian models are within the range expected from different meteorological variations.

# Source-receptor (country-to-country) matrix for oxidized sulphur in 1997

(Units: 100 tonnes of S, Emitters → , Receptors ↓)

AT	BE	BG	DK	FI	FR	DE	GR	HU	IS	IE	IT	LU	NL	NO	PL	PT	RO	ES	SE	CH	TR	GB	BY	UA	MD	RU	LV	LT	CZ	SK	SI	HR	BA	YU*	MK	CY	REM	BAS	NOS	ATL	MED	BLS	NAT	VUL	SUM		
61	13	10	2	0	48	145	3	33	0	1	89	1	5	0	50	1	8	30	0	5	0	18	1	2	0	0	0	78	19	65	6	8	7	0	0	3	2	7	1	0	0	1	32	758	AT		
0	165	0	1	0	76	40	0	1	0	1	3	1	45	0	4	1	0	24	0	0	0	32	0	0	0	0	0	6	0	0	0	0	1	0	0	1	0	24	2	0	0	0	2	3	437	BE	
2	2	1179	0	0	8	32	63	62	0	0	49	0	0	0	55	1	228	18	0	0	4	2	45	2	0	0	21	15	5	4	52	147	7	0	17	1	1	1	0	0	0	0	2	110	2149	BG	
1	9	1	73	0	0	21	49	0	4	1	2	3	0	5	1	28	1	12	4	0	0	44	0	1	0	6	0	7	2	1	0	1	1	0	0	1	34	26	2	0	0	7	2	354	DK		
1	7	2	9	0	201	19	70	0	16	0	1	7	0	3	3	74	0	8	19	19	0	0	24	15	11	0	288	11	9	20	6	2	1	6	6	0	54	55	8	3	0	0	7	13	1003	FI	
4	111	11	4	0	1560	184	6	18	0	10	156	8	74	0	34	30	5	786	1	15	0	174	0	2	0	1	0	0	37	5	9	4	11	8	0	0	22	3	105	53	1	0	25	107	3540	FR	
20	151	8	23	1	353	1711	3	41	0	9	58	8	31	0	315	6	8	149	3	22	0	197	3	4	0	12	2	3	442	15	9	2	10	8	0	0	7	42	94	11	0	0	12	49	3866	DE	
1	1	482	0	0	9	16	437	31	0	0	65	0	0	0	0	26	1	56	31	0	5	2	1	15	0	2	0	0	11	7	3	2	32	58	9	1	39	0	1	1	0	0	4	175	1529	GR	
13	3	28	1	0	13	72	9	596	0	0	51	0	1	0	137	1	99	21	0	1	1	5	2	12	0	4	0	60	92	18	22	49	79	0	0	6	2	2	3	0	0	14	1	53	1459	HU	
0	2	0	0	0	5	3	0	0	22	3	1	0	1	0	0	2	0	9	0	0	0	17	0	0	0	0	0	1	0	0	0	0	0	0	0	0	0	2	0	0	0	0	10	1	90	IS	
0	5	1	1	0	19	8	0	1	0	202	2	0	2	0	2	2	0	35	0	0	0	88	0	0	0	0	0	1	0	0	0	0	0	0	0	1	7	15	0	0	0	10	5	408	IE		
12	9	49	1	0	103	65	29	47	0	1	1641	1	3	0	44	9	25	160	0	9	1	14	6	0	1	0	0	34	11	53	24	72	42	1	0	37	1	5	5	0	0	10	1367	3894	IT		
0	5	0	0	0	8	5	0	0	0	0	0	1	2	1	0	0	0	4	0	0	0	40	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	1	30	LU		
0	68	0	1	0	47	64	0	2	0	2	2	0	108	0	8	1	0	19	0	0	0	40	0	0	0	1	0	7	1	0	0	1	1	43	2	0	0	1	43	2	0	0	3	3	427	NL	
14	4	18	5	40	97	2	10	1	7	6	0	6	49	47	1	4	34	14	1	4	0	100	3	3	0	151	3	3	17	4	2	0	4	4	0	6	18	37	15	0	0	28	8	764	NO		
12	27	53	24	4	68	731	12	181	0	3	49	1	13	2	4909	3	67	55	8	2	1	65	31	69	1	53	9	23	437	91	18	7	36	41	1	0	15	52	24	3	0	0	7	58	7263	PL	
0	0	0	0	0	25	106	52	222	0	1	104	0	3	0	235	2	1452	32	1	1	8	12	10	149	6	21	2	0	0	0	0	0	0	0	3	0	0	0	0	0	0	0	7	175	3525	RO	
3	22	6	57	31	56	181	2	28	0	5	14	1	9	18	113	1	9	43	119	1	0	93	10	9	0	101	10	35	9	4	2	10	9	0	23	114	35	8	0	0	16	14	1232	SE			
2	8	2	0	0	64	23	1	2	0	0	64	1	3	0	4	2	1	42	0	0	39	0	12	0	0	0	0	5	0	3	1	2	1	0	2	0	4	1	0	0	0	1	20	309	CH		
4	3	532	1	0	20	49	142	56	0	0	118	0	1	0	81	1	162	56	0	1	778	9	7	177	3	57	1	2	27	16	9	4	53	74	3	32	318	2	2	1	0	0	16	299	3118	TR	
1	30	4	4	0	107	70	1	5	0	61	12	1	14	1	16	5	3	101	1	1	0	2625	0	1	0	1	0	0	13	2	1	1	4	3	0	3	147	43	0	0	31	16	3331	GB			
3	7	45	7	5	20	121	7	55	0	1	21	0	3	1	388	0	49	19	3	1	2	21	290	110	1	87	20	42	43	21	5	3	19	25	0	20	14	5	1	0	0	2	31	1530	BY		
9	11	325	7	3	35	232	62	252	0	1	107	0	5	1	738	1	449	42	3	1	25	26	104	1806	21	211	9	20	110	87	19	10	85	111	3	1	59	13	7	2	0	0	7	200	5222	UA	
0	1	30	0	0	2	10	5	11	0	0	7	0	0	0	28	0	85	2	0	0	1	3	47	8	4	0	1	5	3	1	6	8	0	0	0	0	0	0	0	0	0	0	0	17	291	MD	
17	41	542	30	104	126	566	109	290	0	7	169	2	15	7	965	0	454	134	26	3	122	106	385	1612	13	7515	89	93	206	98	37	14	127	150	4	7	861	80	28	12	0	0	32	241	15471	RU	
1	4	8	4	5	11	47	2	12	0	1	6	0	2	1	66	0	7	8	3	0	0	11	15	12	0	22	43	26	13	4	1	1	4	4	0	15	16	4	1	0	0	2	8	390	LV		
15	12	7	3	0	34	352	2	72	0	1	21	1	5	0	413	1	11	19	1	1	0	16	2	5	0	4	1	1	1058	37	10	3	9	10	0	0	3	5	7	1	0	0	1	17	2159	CZ	
6	3	19	1	0	9	68	5	172	0	0	23	0	1	0	219	1	27	11	0	0	0	4	2	11	0	4	1	68	133	9	4	20	22	0	0	3	2	2	0	0	0	0	1	25	879	SK	
7	2	17	0	0	15	35	7	72	0	0	71	0	1	0	35	1	19	29	0	1	0	3	0	3	0	1	0	0	25	13	27	102	114	47	0	6	1	1	1	0	0	1	50	708	HR		
4	2	21	1	0	16	35	9	55	0	0	69	0	1	0	33	1	16	26	0	1	1	5	1	4	0	1	0	24	11	8	16	461	60	1	0	9	1	2	1	0	0	1	68	966	BA		
5	4	84	1	0	19	54	26	135	0	0	86	0	1	0	82	1	107	24	0	1	1	7	1	15	0	3	0	1	38	27	9	13	255	655	6	0	2	1	0	0	0	1	114	1803	YU*		
1	0	53	0	0	3	6	32	11	0	0	21	0	0	0	9	0	14	9	0	0	1	1	0	2	0	0	0	4	3	1	1	14	35	13	0	19	0	0	0	0	0	0	0	45	300	MK	
0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	1	2	36	CY
0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	18	0	0	2	0	2	0	0	0	0	0	0	0	0	1	62	0	0	0	0	0	0	0	3	104	AM	
6	9	321	4	11	79	207	83	0	2	365	0	2	0	0	1	121	23	120	266	4	2	257	22	24	299	2	469	16	10	39	22	17	8	94	101	6	35	4131	22	6	21	3	0	45	881	8253	REM
5	212	13	134	90	83	380	3	53	0	6	22	1	3	1	476	1	21	53	93	1	0	104	22	23	0	100	41	33	74	20	7	3	17	16	0	0	81	540	52	6	0	0	28	28	2689	BAS	
NOS	5	212	13	134	90	83	380	3	53	0	6	22	1	3	1	476	1	21	53	93	1	0	104	22	23	0	100	41	33	74	20	7	3	17	16	0	0	81	540	52	6	0	0	28	28	2689	BAS
ATL	7	127	9	33	22	691	423	7	39	93	415	87	4	46	33	155	844	19	2314	16	5	1	1768	14	25	1	1020	7	8	96	14	10	3	8	15	0	0	51	34	237	2560	5	0	2798	136	14200	ATL
MED	23	17	1214	3	0	411	181	810	259	0	2	2142	1	7	0	209	42	254	970	1	6	149	40	5	76	3	11	2	114	60	80	89	388	2													





**Table 1.1: Emissions of sulphur dioxide (100 tonnes as S per year)**

	1980	1985	1986	1987	1988	1989	1990	1991	1992	1993	1994	1995	1996	1997	2000	2005	2010
Africa, north1	2065	2065	2065	2065	2065	2065	2065	2065	2065	2065	2065	2065	2065	2065	2065	2065	2065
Albania	360	360	360	360	360	360	360	360	360	360	360	360	360	360	360	360	360
Armenia	555	500	555	555	520	315	360	300	220	30	20	15	10	2	2	2	2
Austria	2000	975	880	800	575	510	455	415	315	300	280	260	260	285	300	300	300
Belarus	3700	3450	3450	3805	3600	3340	3185	3260	2290	1910	1620	1375	1230	1040	2760	2450	2400
Belgium	4140	2000	1885	1835	1770	1625	1610	1630	1550	1445	1255	1235	1200	1200	1240	1160	1075
Bosnia and Herzegovina	2400	2400	2400	2400	2400	2400	2400	2400	2400	2400	2400	2400	2400	2400	2400	2400	2400
Bulgaria	10250	11570	11835	12100	11140	10900	10100	8385	5640	7130	7400	7485	7100	6825	6870	6150	5635
Croatia	750	825	840	855	870	885	900	540	535	570	445	340	360	400	665	625	585
Cyprus	185	185	185	205	215	235	275	205	225	215	230	230	230	235	215	185	195
Czech Republic	11285	11385	10885	10820	10330	9990	9380	8880	7690	7095	6350	5455	4730	3505	2310	2105	1880
Denmark	2250	1695	1410	1250	1210	955	910	1215	950	780	775	750	930	545	450	450	450
Estonia	1435	1270	1280	1275	1270	1270	1260	1230	935	770	745	590	625	595	1260	1260	1260
Finland	2920	1910	1655	1640	1510	1220	1300	970	705	620	560	480	525	500	580	580	580
France	16690	7350	6710	6450	6130	6670	6490	6880	6190	5605	5065	4945	5155	5155	4340	3850	3685
Georgia	810	810	810	810	810	810	810	810	810	810	810	810	810	810	810	810	810
Germany, former East	21750	26825	27065	27215	26315	26270	22130	15625	12305	10620	9210	7590	5570	5300	4695	3575	1985
Germany, former West	15820	11835	11140	9520	6075	4710	4435	4355	4190	4070	3120	2920	2145	2040	1805	1375	765
Greece	2000	2500	2510	2520	2525	2535	2545	2760	2770	2750	2690	2765	2715	2715	2975	2900	2850
Hungary	8165	7020	6810	6425	6090	5510	5050	4565	4135	3810	3705	3525	3365	3285	4490	4080	3265
Iceland	90	90	90	80	90	85	120	115	120	125	120	120	120	125	145	145	150
Ireland	1110	700	810	870	760	810	890	895	805	785	885	805	735	825	775	775	775
Italy	18785	9505	9645	10145	9815	9270	8255	7695	6970	6665	6355	6610	6610	6610	5020	4235	4210
Kazakhstan1	700	700	700	700	700	700	700	700	700	700	700	700	700	700	700	700	700
Latvia	595	595	595	595	595	595	595	450	395	365	430	295	295	295	385	570	785
Lithuania	1555	1520	1580	1580	1500	1490	1110	1170	695	625	585	470	465	385	820	775	725
Luxembourg	120	85	85	80	80	75	75	75	70	70	65	45	40	40	20	20	20
Netherlands	2450	1290	1320	1315	1250	1020	1010	865	860	820	730	735	675	620	460	460	490
Norway	685	490	455	365	340	290	265	220	180	175	170	170	165	150	170	170	170
Poland	20500	21500	21000	21000	20900	19550	16050	14975	14100	13625	13025	11880	11840	10905	16050	16050	16050
Portugal	1330	990	1155	1320	1480	1645	1810	1760	2100	1780	1680	1865	1865	1865	1520	1470	1470
Republic of Moldova	1540	1410	1485	1585	1365	1190	1155	920	630	180	115	135	95	85	700	650	650
Romania	5275	6275	6465	6525	7345	7585	6555	5205	4755	4640	4560	4560	4560	4560	6555	6555	6555
Russian Federation1	35805	30955	28535	28110	25725	23385	22300	21960	19195	17280	14915	14190	13425	12245	22200	21485	21485
Slovakia	3900	3065	3020	3070	2945	2865	2715	2230	1900	1625	1195	1195	1135	1010	1050	1050	1050
Slovenia	1170	1205	1235	1110	1050	1060	970	905	950	915	885	595	550	600	460	225	185
Spain2	16595	10950	9805	9515	7935	9750	11330	11115	10975	10355	10305	10305	10305	10305	10715	10715	10715
Sweden	2455	1330	1360	1140	1120	800	595	480	440	435	410	395	415	345	360	335	335
Switzerland	580	380	340	310	280	245	215	205	190	170	155	170	150	130	130	130	135
The FYR Macedonia	85	85	85	85	85	85	85	85	85	85	85	85	85	85	85	85	85
Turkey	4300	1610	1770	1770	1770	1770	1770	1770	1770	1770	1770	1770	1770	1770	1770	1770	1770
Ukraine	19245	17315	16965	16320	16055	15365	13910	12690	11880	10970	8575	8195	6465	5660	11550	11550	11550
United Kingdom	24310	18645	19500	19435	19080	18465	18655	17740	17280	15695	13395	11715	10085	8280	6450	5100	4250
Yugoslavia	2030	2390	2350	2420	2510	2530	2540	2230	1980	2005	2120	2310	2170	2610	3400	4445	5675
Other Asiatic areas1	4345	4345	4345	4345	4345	4345	4345	4345	4345	4345	4345	4345	4345	4345	4345	4345	4345
The Baltic Sea	1140	1140	1140	1140	1140	1140	1140	1140	1140	1140	1140	1140	1140	1140	1140	1140	1140
The Mediterranean Sea	60	60	60	60	60	60	60	60	60	60	60	60	60	60	60	60	60
The North Sea	2270	2270	2270	2270	2270	2270	2270	2270	2270	2270	2270	2270	2270	2270	2270	2270	2270
Rem. N.E. Atlantic1	4505	4505	4505	4505	4505	4505	4505	4505	4505	4505	4505	4505	4505	4505	4505	4505	4505
Natural Oceanic1	3715	3715	3715	3715	3715	3715	3715	3715	3715	3715	3715	3715	3715	3715	3715	3715	3715
Volcanic3	10720	10720	10720	10905	10565	12465	14035	8225	11175	10135	9590	10000	10000	10000	10000	10000	10000
Total	301495	256765	251835	249290	237155	231700	219765	197565	182515	171385	157960	150945	142540	134490	158115	152180	148565

**Table 1.2: Emissions of nitrogen oxides (100 tonnes as N per year)**

	1980	1985	1986	1987	1988	1989	1990	1991	1992	1993	1994	1995	1996	1997	2000	2005	2010
Africa, north1	292	292	292	292	292	292	292	292	292	292	292	292	292	292	292	292	292
Albania	73	73	73	73	73	73	73	73	73	73	73	73	73	73	73	73	73
Armenia	46	137	161	158	170	155	140	122	67	37	37	46	33	46	140	140	140
Austria	703	670	660	648	618	597	590	603	572	536	560	517	496	523	469	469	469
Belarus	712	724	785	800	797	800	867	855	682	630	618	593	527	575	660	560	548
Belgium	1345	989	965	1029	1050	1087	1044	1047	1077	1047	1053	1032	1017	1017	1044	1044	1044
Bosnia and Herzegovina	243	243	243	243	243	243	243	243	243	243	243	243	243	243	243	243	243
Bulgaria	1266	1266	1266	1266	1263	1251	1144	810	727	737	700	810	788	685	1157	1065	883
Croatia	265	265	265	265	265	265	265	195	170	180	198	195	204	225	265	253	253
Cyprus	49	49	49	49	55	58	61	52	61	64	64	64	64	70	64	70	70
Czech Republic	2852	2529	2514	2483	2611	2800	2258	2207	2124	1747	1324	1254	1315	1287	1132	1102	1068
Denmark	858	916	974	950	919	858	858	977	840	834	828	761	877	755	618	584	584
Estonia	213	213	213	213	213	210	207	192	119	116	128	128	134	137	207	207	207
Finland	898	837	843	877	892	916	913	883	861	858	858	788	813	791	682	682	682
France	5548	4915	4924	4961	4915	5393	4824	4967	4860	4699	5119	5070	4994	4994	4824	4824	4824
Georgia	572	572	572	572	572	572	572	572	572	572	572	572	572	572	572	572	572
Germany, former East	2182	2240	2252	2283	2261	2270	2130	1802	1537	1446	1284	1196	1160	1108	2130	1309	776
Germany, former West	7965	7730	7749	7387	7018	6531	6066	5871	5487	5101	4863	4727	4583	4380	6066	5174	3068
Greece	931	931	953	977	998	1023	1044	1074	1071	1062	1090	1090	1138	1138	1044	1044	1044
Hungary	831	797	803	807	785	749	724	618	557	560	572	578	597	603	700	639	597
Iceland	64	64	67	73	76	76	79	82	85	88	88	85	91	88	85	88	91
Ireland	222	277	304	350	371	387	350	362	380	371	356	350	368	377	320	320	320
Italy	4985	4912	5143	5512	5643	5834	5898	6038	6117	6057	5445	5381	5381	5381	4930	4590	4370
Kazakhstan1	231	231	231	231	231	231	231	231	231	231	231	231	231	231	231	231	231
Latvia	283	283	283	283	283	283	283	186	161	140	146	128	107	107	170	213	247
Lithuania	463	505	514	520	523	527	481	505	298	237	234	198	198	173	332	335	335
Luxembourg	70	64	64	67	67	70	70	70	70	70	70	61	67	67	58	58	58
Netherlands	1774	1793	1787	1823	1832	1777	1765	1729	1692	1628	1552	1516	1525	1430	1765	758	995
Norway	572	639	688	685	673	670	663	633	630	654	645	645	670	676	481	481	481
Poland	3740	4565	4596	4657	4717	4504	3896	3667	3439	3409	3363	3409	3512	3524	2453	2453	2675
Portugal	292	292	444	600	752	907	1059	1117	1190	1163	1190	1239	1239	1239	1059	1059	1059
Republic of Moldova	177	204	219	216	225	213	192	204	119	46	55	73	33	91	91	107	103
Romania	1592	1650	1701	1765	1796	1762	1662	1412	1087	968	971	971	971	971	1662	1662	1662
Russian Federation1	5277	5792	5694	8074	7177	7770	10957	10120	9413	9295	8172	7822	7508	7240	10957	10957	10957
Slovakia	600	600	600	600	645	691	685	645	578	557	527	551	396	374	685	685	685
Slovenia	155	161	177	173	180	177	189	164	167	186	201	204	213	216	137	116	94
Spain	2891	2553	2599	2715	2715	3019	3582	3734	3807	3722	3722	3722	3722	3722	2715	2715	2715
Sweden	1230	1297	1315	1330	1315	1272	1029	1032	1001	986	1007	916	919	852	831	633	609
Switzerland	517	545	539	530	523	517	505	487	466	441	423	414	396	380	356	335	344
The FYR Macedonia	18	18	18	18	18	18	18	18	18	18	18	18	18	18	18	18	18
Turkey	837	1108	1205	1297	1254	1412	1513	1558	1628	1710	1908	2106	2106	2106	2907	3731	5083
Ukraine	3485	3223	3384	3330	3317	3241	3339	3010	2526	2130	1729	1616	1421	1385	3330	3330	3330
United Kingdom	7642	7405	7645	7968	8141	8278	8175	7864	7639	7137	6854	6403	6175	5624	4739	3941	3533
Yugoslavia	143	177	177	183	192	189	201	173	149	164	158	180	173	201	268	350	447
Other Asiatic areas1	645	645	645	645	645	645	645	645	645	645	645	645	645	645	645	645	645
The Baltic Sea	1071	1071	1071	1071	1071	1071	1071	1071	1071	1071	1071	1071	1071	1071	1071	1071	1071
The Mediterranean Sea	40	40	40	40	40	40	40	40	40	40	40	40	40	40	40	40	40
The North Sea	1972	1972	1972	1972	1972	1972	1972	1972	1972	1972	1972	1972	1972	1972	1972	1972	1972
Rem. N.E. Atlantic1	3853	3853	3853	3853	3853	3853	3853	3853	3853	3853	3853	3853	3853	3853	3853	3853	3853
Natural Oceanic	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0
Volcanic	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0
Total	72687	72328	73533	76912	76260	77551	78720	76078	72471	69823	67124	65849	64942	63572	70542	67090	65459

**Table 1.3: Emissions of ammonia (100 tonnes as N per year)**

	1980	1985	1986	1987	1988	1989	1990	1991	1992	1993	1994	1995	1996	1997	2000	2005	2010
Africa, north1	1935	1935	1935	1935	1935	1935	1935	1935	1935	1935	1935	1935	1935	1935	1935	1935	1935
Albania	255	255	255	255	255	255	255	255	255	255	255	255	255	255	255	255	255
Armenia	25	16	16	16	16	1.6	0.5	0.9	0.4	0.08	0.05	0.05	0.03	0.03	0.5	0.5	0.5
Austria	659	675	675	667	667	642	626	601	609	618	642	642	626	618	626	626	626
Belarus	1804	1804	1804	1804	1804	1804	1804	1804	1804	1804	1804	1804	1804	1804	1804	1804	1804
Belgium	733	733	758	782	807	832	856	766	758	799	791	799	799	799	856	856	856
Bosnia and Herzegovina	255	255	255	255	255	255	255	255	255	255	255	255	255	255	255	255	255
Bulgaria	1186	1186	1186	1186	1186	1186	1186	1021	914	898	832	815	684	634	898	1038	1038
Croatia	305	305	305	305	305	305	305	255	222	206	198	206	189	198	272	272	272
Cyprus	33	33	33	33	33	33	33	33	33	33	33	33	33	33	33	33	33
Czech Republic	1285	1285	1285	1285	1285	1285	1285	1104	947	815	749	708	667	667	1285	1285	1285
Denmark	1161	1038	1005	964	947	931	1005	906	939	980	931	939	815	840	848	848	848
Estonia	239	239	239	239	239	239	239	239	239	239	239	239	239	239	239	239	239
Finland	288	288	288	288	288	288	288	288	288	288	288	288	288	280	264	189	189
France	5765	5765	5765	5765	5765	5765	5765	5682	5567	5485	5501	5501	5501	5501	5765	5765	5765
Georgia	799	799	799	799	799	799	799	799	799	799	799	799	799	799	799	799	799
Germany, former East	2166	2215	2191	2182	2224	2158	1738	1145	1038	1013	1021	1046	1046	1054	1738	1738	1738
Germany, former West	4711	4842	4776	4711	4612	4579	4554	4422	4356	4307	4332	4274	4299	4282	4554	4554	4554
Greece	881	881	881	881	881	881	881	881	881	881	881	881	881	881	881	881	881
Hungary	1614	1556	1515	1474	1433	1392	1351	1104	1013	947	939	955	642	626	1400	1318	1235
Iceland	25	25	25	25	25	25	25	25	25	25	25	25	25	25	25	25	25
Ireland	1038	1038	1038	1038	1038	1038	1038	1038	1038	1038	1013	1021	1054	1087	1038	1038	1038
Italy	3945	4011	4076	4093	4109	3961	3838	3714	3624	3698	3780	3796	3796	3796	3648	3673	3698
Kazakhstan1	148	148	148	148	148	148	148	148	148	148	148	148	148	148	148	148	148
Latvia	362	362	362	362	362	362	362	346	272	165	140	140	140	140	362	362	362
Lithuania	700	733	733	741	733	708	692	700	667	659	659	313	296	288	692	692	692
Luxembourg	58	58	58	58	58	58	58	58	58	58	58	58	58	58	49	49	49
Netherlands	1927	2042	2125	2125	1952	1911	1861	1878	1482	1573	1367	1202	1202	1194	1194	675	1120
Norway	189	189	189	189	173	189	189	198	206	206	206	214	222	214	189	189	189
Poland	4529	4529	4529	4529	4529	4529	4184	3706	2816	3146	3162	3129	2998	2882	4184	4184	4184
Portugal	807	807	807	807	807	807	807	807	791	774	774	799	799	799	807	807	807
Republic of Moldova	387	387	387	387	387	387	387	387	387	387	387	387	387	387	387	387	387
Romania	2800	2825	2882	2709	2792	2808	2471	2199	2100	1836	1820	1820	1820	1820	2471	2471	2471
Russian Federation1	9792	10204	10591	10516	10451	10360	9808	9561	8927	7436	6358	6786	6168	6012	9808	9808	9808
Slovakia	511	511	511	511	511	511	511	511	502	420	329	371	412	412	511	511	511
Slovenia	198	198	198	198	198	198	198	198	189	189	181	181	181	181	222	222	222
Spain	2907	2907	2907	2907	2907	2907	2907	2915	2899	2841	2833	2833	2833	2833	2907	2907	2907
Sweden	445	445	445	445	445	436	420	420	502	502	502	502	502	527	527	395	395
Switzerland	634	609	609	601	601	593	593	593	593	585	585	585	585	585	576	568	560
The FYR Macedonia	140	140	140	140	140	140	140	140	140	140	140	140	140	140	140	140	140
Turkey	2644	2644	2644	2644	2644	2644	2644	2644	2644	2644	2644	2644	2644	2644	2644	2644	2644
Ukraine	6004	6004	6004	6004	6004	6004	6004	6004	6004	6004	6004	6004	6004	6004	6004	6004	6004
United Kingdom	2742	2742	2742	2742	2742	2742	2742	2726	2701	2685	2668	2644	2627	2660	2742	2742	2742
Yugoslavia	741	741	741	741	741	741	741	741	741	741	741	741	741	741	741	741	741
Other Asiatic areas1	2495	2495	2495	2495	2495	2495	2495	2495	2495	2495	2495	2495	2495	2495	2495	2495	2495
The Baltic Sea	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0
The Mediterranean Sea	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0
The North Sea	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0
Rem. N.E. Atlantic1	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0
Natural Oceanic	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0
Volcanic	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0
Total	72265	72899	73352	72981	72726	72265	70420	67645	64804	62951	61444	61353	60035	59772	69218	68567	68946

## *Overview of Reported Emissions*



## ***2. Overview of EMEP reported emission data***

Since the start of the LRTAP Convention, the improvement in emission reporting to EMEP has been massive, aided greatly by the establishment of the relevant guidelines. Emission reporting has improved markedly since last year, specially concerning national and sectoral totals. However, Parties show still difficulties on reporting reporting gridded data and projections within the established deadlines.

Following the recommendations from the last meeting of the Task Force on Emissions Inventories in June 1999, this chapter reviews the EMEP emission reporting requirements and evaluates them in relation to the main objectives of the acidification programme for its seventh phase. A complete report on the present status of the UN/ECE-EMEP emission data and the emission data used for modilling purposes at MSC-W is given in Mylona (1999). Updated emission values up to 1997 will be available on the Internet at <http://www.emep.int> from mid-September 1999.

### ***2.1 Main objectives for the EMEP emission data***

Under the Convention on Long Range Transboundary Air Pollution (LRTAP) the supply of good quality emission data is essential in:

- a) assessing the state and trends of air pollution in Europe,
- b) establishing the compliance of the Parties with protocol commitments, and
- c) providing a basis for development of cost-effective abatement strategies,

These are the main objectives for the work of EMEP during its seventh phase. The quality of the emission data is measured in terms of its:

- completeness,
- transparency
- consistency
- comparability
- accuracy

The assessment of trends in emissions and depositions to survey the progress towards environmental goals imposes an increased demand for completeness, consistency and transparency in the emission data. The completeness of the data concerns both the temporal evolution and the geographical distribution.

The review of compliance with international agreements for emission reductions imposes strong requirements on the accuracy of emission values and an increased need for the evaluation of uncertainties and verification of the emission data.

The evaluation of the effects of specific control measures requires transparent and consistent and emissions inventories where the different contributions to total emissions can be separated. The requirement of transparency and comparability is specially important when national inventories are aggregated to broader geographical areas, like in the case of EMEP, and the need for international harmonisation becomes then significant.

In this sense, emission reporting to UN/ECE-EMEP is greatly assisted by the Task Force of Emission Inventories which is an international forum for technical exchange of information and was established to harmonise methodologies in emission estimation and reporting. An important tool developed for this purpose is Joint EMEP/CORINAIR Atmospheric Emission Inventory Guidebook.

## ***2.2. EMEP emission reporting requirements***

The objectives for the EMEP emission data specified above translate into concrete operational requirements on the type of data and reporting that should be submitted to UN/ECE-EMEP. The requirements for estimating and reporting emission data under the CLRTAP are presently drafted for consideration by the countries in EB.AIR/GE.1/1997/2.

During the last meeting of the Task Force on Emission Inventory, the draft reporting procedures were discussed and reviewed. It was agreed the reporting procedures could be revised in order to increase their clarity. Task Force members were invited to provide their comments to MSC-W and to the UNECE Secretariat and new revised guidelines would be circulated for comments. In order to stimulate these comments, Table 2.1 summarises the reporting requirements, as they are interpreted by MSC-W.

It has been suggested that the emission data requirements as they appear in Table 2.1 may not be sufficient to secure that the EMEP programme can reach its objectives. The main concern is on the transparency of the emission data and SNAP level 2 has been suggested as the necessary level of disaggregation of sectoral data for EMEP assessment purposes. Studies on the effects of different reduction strategies will profit from the disaggregation of emission data to SNAP level 2.

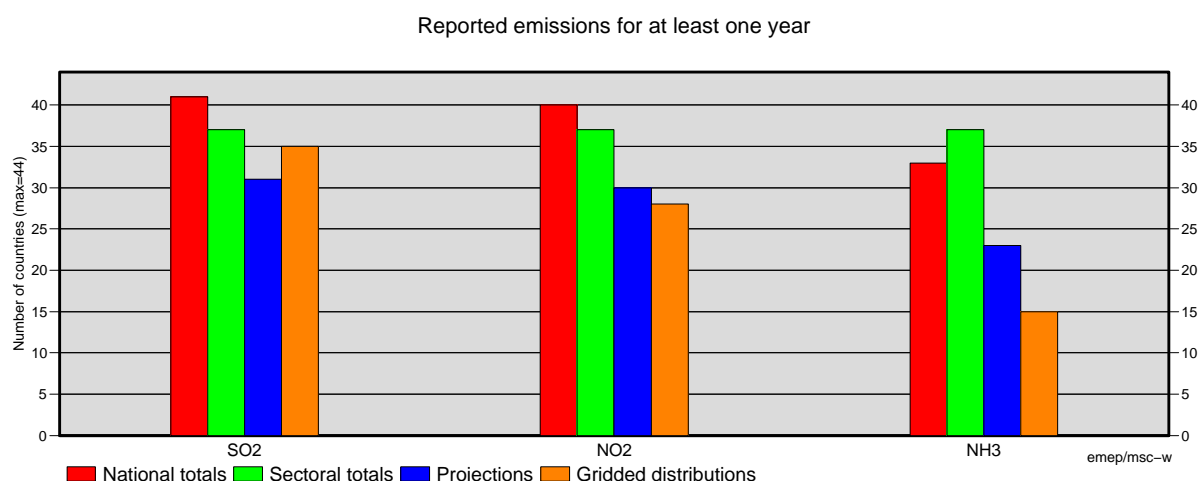
Another suggestion is to report the height distribution of emissions. The actual height of sources will be most useful for atmospheric modelling, which at present can discern better different atmospheric levels than what did in the past. With the use of Eulerian dispersion models, information on the height distribution of emissions has become increasingly important and needs to be completed or updated as soon as possible. Information on the position and characteristics of LPS is extremely useful in shedding light on issues related to both emission patterns, monitoring data and model results.

**Table 2.1** Emission reporting requirements for acidification, as interpreted by MSC-W\_

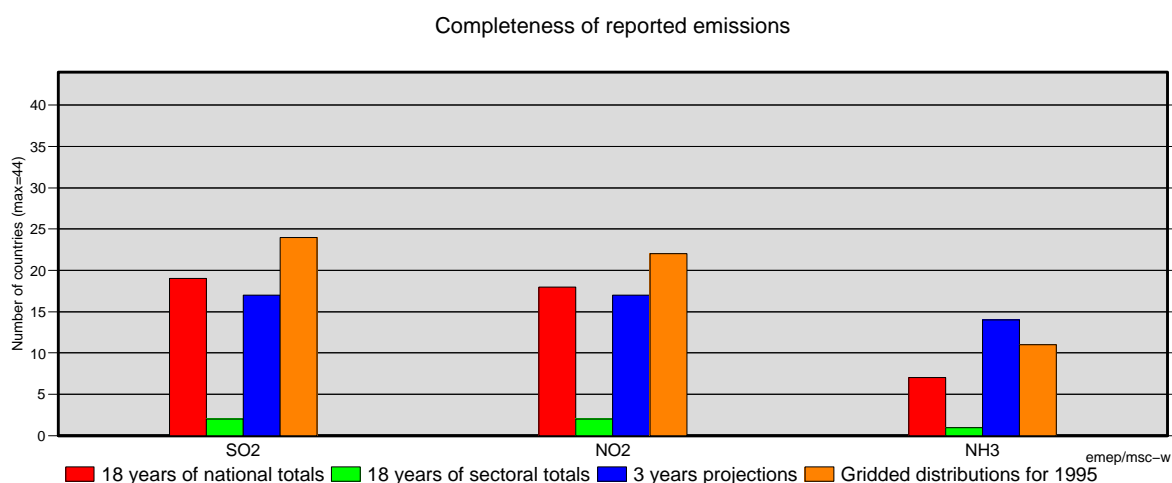
<b>Emission reporting requirements</b>						
Emissions of SO <sub>2</sub> , NO <sub>x</sub> and NH <sub>3</sub>	Reporting frequency	Temporal resolution	Spatial resolution	SNAP level	Methods	Comments
<b>General reporting:</b>						
National annual totals	1 year	annual	-	-	ECM*	
Sectoral emissions at SNAP level 1	1 year	annual	-	1 (11 sectors)	ECM*	
Emissions projections for 2000,2005,2010	1 year	annual	-	1 (11 sectors)	ECM*	
National totals from low and high sources	1 year	annual	-	1 (11 sectors)	ECM*	
<b>Specific reporting for modelling:</b>						
Spatial distribution of annual totals	5 years	annual	50x50 km <sup>2</sup> *	-	ECM*	SCRA*
Spatial distribution of sectoral emissions	5 years	annual	50x50 km <sup>2</sup> *	1 (11 sectors)	ECM*	SCRA*
Spatial distribution of low sources (<100m)	5 years	annual	50x50 km <sup>2</sup> *	1 (11 sectors)	ECM*	SCRA*
Spatial distribution of high sources (>100m)	5 years	annual	50x50 km <sup>2</sup> *	1 (11 sectors)	ECM*	SCRA*
Spatial distribution of large point sources	5 years	annual	50x50 km <sup>2</sup> *	1 (11 sectors)	ECM*	SCRA*
<b>Extended reporting:</b>						
National annual totals	5 years	monthly	-	-		
Sectoral emissions	5 years	monthly	50x50 km <sup>2</sup> *	1(11 sectors)	ECM*	
Large Point sources	5 years	monthly	50x50 km <sup>2</sup> *	1 (11 sectors)	ECM*	Stack heightI
<b>Comments:</b>						
<p><b>50x50 km<sup>2</sup>* : gridded data in the EMEP 50x50 km<sup>2</sup> polar stereographic grid,</b>  <b>ECM* : EMEP/CORINAIR Manual methods : differences in methodology used should be documented</b>  <b>SCRA* : substantial changes in emission distribution should be reported annually</b></p>						

### 2.3. Present status (1997 emission data)

A major objective in the EMEP emission inventorying work is to obtain complete time series of national annual total and sectoral emissions since 1980 using harmonised emission inventory methodologies, as well as full sets of gridded national and sectoral data for every five years starting from 1980 and updates in between. These targets should be reached as soon as possible and at the latest during the 2005 inventory.



**Figure 2.1** Number of Parties having reported national totals, sectoral totals, emission projections and gridded distributions for at least one year.



**Figure 2.2** Number of Parties having reported national totals, sectoral totals and emission projections for all requested years, and gridded distributions after 1995.

The completeness of the emission data is essential for the determination of trends. Figures 2.1 and 2.2 show the 1997 status with respect to completeness. While national total emissions for the major components have been reported for at least one year by more than 2/3 out of total 44 Parties, only about 40% of the Parties have reported all years since 1980. For ammonia, only 16% of the Parties has reported 18 national emission totals. For national sectoral emissions, the

completeness of the data is not satisfactory, as only 4% of the Parties have reported sectoral national totals for all years.

Keeping in mind the objectives for the seventh phase of EMEP, the following suggestions for improvements can be made:

*1) Increase the consistency of reported data, specially concerning the sectoral data*

Concerning the officially reported sectoral data, a major observation to be stressed is that Parties revising their total emissions for earlier years, rarely do so for the respective sectoral totals. In several cases revisions of national totals are considerable and update of sectoral totals can by no means be overlooked. Reporting on the emission estimation methodology followed is necessary to aid work on quality control and validation of the reported figures.

*2) Target for completeness of the reported emissions*

The major objectives in the EMEP emission inventorying work are to obtain complete time series of national annual total and sectoral emissions since 1980 using harmonised emission inventory methodologies, as well as full sets of gridded national and sectoral data for every five years starting from 1980 and updates in between. These targets should be reached as soon as possible and at the latest during the 2005 inventory.

*3) Increase the effort for evaluating emission projections*

*4) Increase efforts for verification of emission data*



## *Overview of Monitoring Network*



### ***3. EMEP monitoring network for acidification and eutrophication***

This chapter presents the 1997 status of the EMEP monitoring network for acidification and eutrophication. The present status of EMEP monitoring network is discussed in relation to its envisaged goals and purposes, providing suggestions for the future development of the programme.

Particular attention is given to clarify the present requirements of the EMEP network for acidification and eutrophication. General specifications in this chapter are complemented with the detailed evaluation of air and precipitation data for each country reported in the Appendix. This evaluation of country contributions to the EMEP network is intended to stimulate the communication between the countries and the EMEP centres. Reactions and comments are both welcome and encouraged.

The following review shows that the present status of the EMEP network is satisfactory for precipitation data but not for the air data programme, where all required nitrogen components were not measured in 1997 at 60% of the stations. Recommendations for the future indicate the desirability of extending the EMEP networks to make them operational for control and surveillance purposes.

#### ***3.1 Main objectives***

The main objective of EMEP is to provide an overview of the European situation with respect to pollution transport across national boundaries. EMEP should provide scientific support for the analysis of the relationships between emissions, atmospheric processes, air concentrations and deposition in a multi-disciplinary programme involving measurements, meteorological modelling and evaluation of effects.

The main goals for the acidification and eutrophication programme are:

- Determination of deposition fluxes for assessment of effects.
- Quantification of transboundary fluxes and source allocation of estimated or measured deposition fluxes.
- Surveillance of the progress towards the reduction of deposition fluxes, in particular as they concern exceedances of critical loads.
- Verification of the reductions of sulphur and nitrogen emissions as prescribed by the Protocols to the CLRTAP.

The requirements to fulfil these goals form the operational strategy of the EMEP programme which has been established by the Steering Body. It is beyond the purpose of this chapter to discuss the operational objectives of EMEP (see instead CCC's note on Strategic Plan for Year 2000, in preparation). In the following, we review the present status of EMEP monitoring network in relation to the existing monitoring requirements and provide suggestions for the future development of the programme.

### ***3.2. EMEP monitoring requirements***

The present operational objectives of EMEP impose a set of specific recommendations for the monitoring programme on acidification and eutrophication, concerning:

- 1) the geographical coverage of the network,
- 2) the component spectrum,
- 3) the temporal coverage and sampling frequency,
- 4) the analytical methods, and
- 5) the QA/QC procedures

Measurement siting criteria are given in the Manual (EMEP/CCC-Report 1/95). The annual data reports contain overview of the site locations and point out areas where sites are missing and where the station densities are low (e.g. Hjellbrekke, 1999). A pilot study on site representativeness is under preparation but a comprehensive survey of EMEP site representativeness is needed. Requirements on the chemical components, temporal coverage, sampling frequency and sampling and analytical methods are summarised in Table 3.1.

It is important to note that the present acidification and eutrophication programs consists of 12 components/parameters in precipitation and 7 mandatory components in air. Detailed descriptions of EMEP recommended methods for sampling and analysis have been presented in two manuals, most recently in the EMEP/CCC-Report 1/95 from 1996. Quality analysis and quality control procedures are documented and discussed in Schaug et al. (1998) and Aas et al. (1999). These procedures include also field comparisons and annual laboratory comparisons of analytical methods (e.g. Hanssen and Skjelmoen, 1997).

The recommendations listed above aim to guarantee a minimum accuracy in the determination of deposition fluxes and their source attribution. This minimum accuracy, together with complete and consistent time series of measurements, are also required for the appropriate compliance monitoring and surveillance of the progress towards the reduction of environmental damages. The requirements on site distribution imposed by compliance monitoring are not clear yet and need further discussion (see Chapter 4).

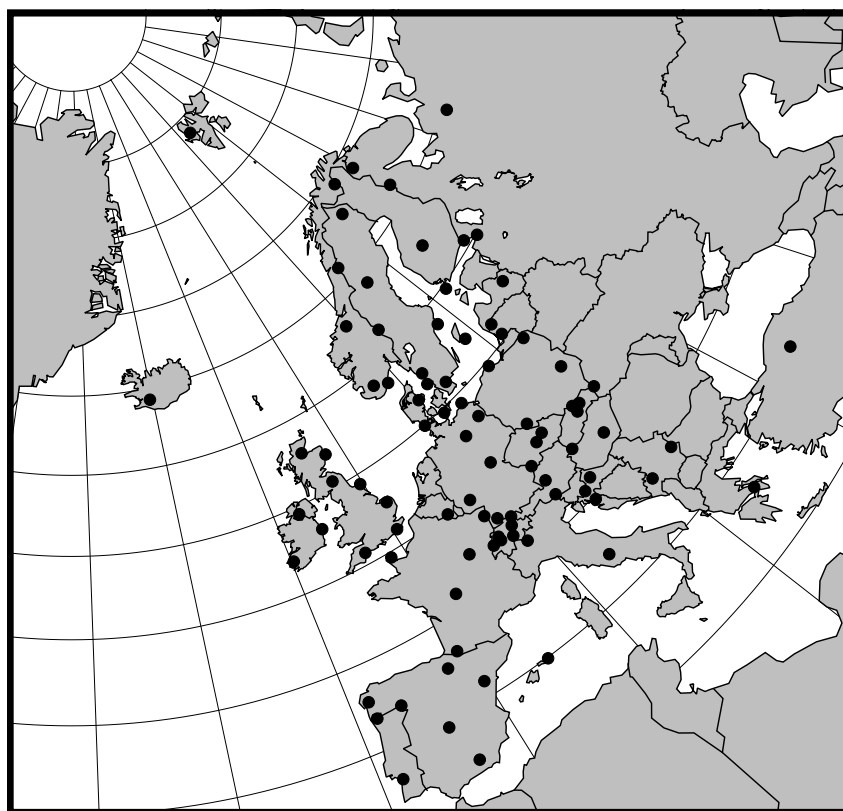
It has been discussed whether weekly sampling could substitute daily sampling in EMEP. For the precipitation data, the present accuracy could probably be also reached by use of wet-only samplers, weekly measurements, parallel sampling with rain gauge, and strict QA protocols. However, for the air data program, daily sampling is necessary. An important goal of EMEP is to provide deposition fluxes for the study of effects. The impact of episodes of combined pollutants (SO<sub>2</sub>, NO<sub>x</sub>, ozone) in different ecosystems is not yet fully understood. Further studies should proceed in this direction, as the focus of the Convention centres in multi-pollutant, multi-effect approaches.

**Table 3.1.** EMEP monitoring requirements for acidification programme

<b>EMEP Acidification Programme requirements</b>					
Component	Sampling frequency	Sampling period	Methods in field	Methods in laboratory	Comments
<b>Precipitation:</b>					
amount	daily	24 hours	by weight		From a rain gauge
SO <sub>4</sub>	daily	24 hours	wet-only	IC	
H	daily	24 hours	wet-only	titration	
pH	daily	24 hours	wet-only	pH meter	
NH <sub>4</sub>	daily	24 hours	wet-only	IC/Indophenol	
NO <sub>3</sub>	daily	24 hours	wet-only	IC/Griess	
Na	daily	24 hours	wet-only	IC/AES	
Mg	daily	24 hours	wet-only	IC/AAS	
Cl	daily	24 hours	wet-only	IC/Thiocyanate	
Ca	daily	24 hours	wet-only	IC/AAS	
K	daily	24 hours	wet-only	IC/AES	
κ	daily	24 hours	wet-only	Cond. meter	
<b>Air:</b>					
SO <sub>2</sub> (g)	daily	24 hours	filter-3-pack	IC	
NO <sub>2</sub> (g)	daily	24 hours	KI-imp glass frit	IC/Griess	
HNO <sub>3</sub> (g)	daily	24 hours	denuder	IC/Griess	
NH <sub>3</sub> (g)	daily	24 hours	denuder	IC/Indophenol	
SO <sub>4</sub> (p)	daily	24 hours	filter-3-pack	IC	
NO <sub>3</sub> (p)	daily	24 hours	denuder	IC/Griess	
NH <sub>4</sub> (p)	daily	24 hours	denuder	IC/Indophenol	
HNO <sub>3</sub> (g)+NO <sub>3</sub> (p)	daily	24 hours	filter-3-pack	IC/Griess	
NH <sub>3</sub> (g)+NH <sub>4</sub> (p)	daily	24 hours	filter-3-pack	IC/Indophenol	
<b>Comments:</b>					
<b>IC: ion chromatography, AES: atomic emission spectroscopy, AAS: atomic absorption spectroscopy</b> <b>filter-3-pack: aerosol filter - KOH impregnated filter oxalic acid impregnated filter</b>					

### 3.3. Present status

The present monitoring strategy in EMEP has resulted in the geographical distribution of EMEP stations illustrated in Figure 3.1.



**Figure 3.1** Geographical coverage of EMEP acidification and eutrophication monitoring network in 1997.

It can be seen that there is a lack of stations in Eastern countries like Bulgaria, Romania, Republic of Moldova, Ukraine, Armenia, Georgia and Russian Federation. The number of stations in Mediterranean countries is also small.

Figure 3.1 can be misleading about the geographical representativeness and completeness of the EMEP programme. In fact, not all stations measure all components requested by the EMEP acidification and eutrophication monitoring program and the analytical and sampling methods used are of very different quality.

Although good progress has been made over the years, full harmonisation of methods has not been achieved. Most countries do not use the recommended methods, and in many cases outdated methods are still in use. The use of a different methodology creates a comparability problem within the network and a need for comparisons between reference and national methods. Although it has been requested that laboratories using other methods than the recommended “should be responsible to document to the CCC their accuracy, precision and comparability to the methods described in the Manual” (EMEP Workshop on data handling, analysis and quality assurance, 1986), this practice is very seldom followed.

The CCC, in collaboration with MSC-W, has recently performed an evaluation of the methods and data quality of EMEP air and precipitation measurements. The evaluation provides an estimate of the expected errors in the 1997 measured annual averages that can be related to the existing sampling and analysis methods. Data from other years than 1997 may have different qualities than those estimated below. It is also expected that errors in single measurements will be larger than in the yearly averages, because random errors in single measurements may cancel each other when data are aggregated. The yearly averages, however, give a good estimate of the existing systematic errors and this is the reason why they have been selected in the evaluation.

The 1997 yearly averages have been classified in four quality groups:

- A < 10%:** expected error 10% or better
- B < 25%:** expected error 25% or better
- C < 30%:** expected error 30% or better
- D > 30%:** expected error worse than 30% or unknown/not documented.

The classification has been based on information from three different sources: a) results from the sixteenth intercomparison of analytical methods within EMEP (Hanssen and Skjelmoen, 1997), b) results from field comparisons contained in (EMEP/CCC-Report 6/98) as well as more recent results from comparisons in Ireland, United Kingdom, Portugal, Germany, France, Poland, and the Czech Republic, and c) calculations of ion balances. Detailed description of the uncertainty estimates can be found in Aas et al., (1999).

Figures 3.2 and 3.3. give an overview of the 1997 data quality for EMEP precipitation and air measurement networks. The figures show the total number of stations for each component included in the EMEP program that have qualified to categories A, B, C, and D. The main conclusions are summarised below and concrete comments for each individual country are given in the Appendix.

- ***The 1997 quality of the EMEP precipitation measurement program is mostly satisfactory.***

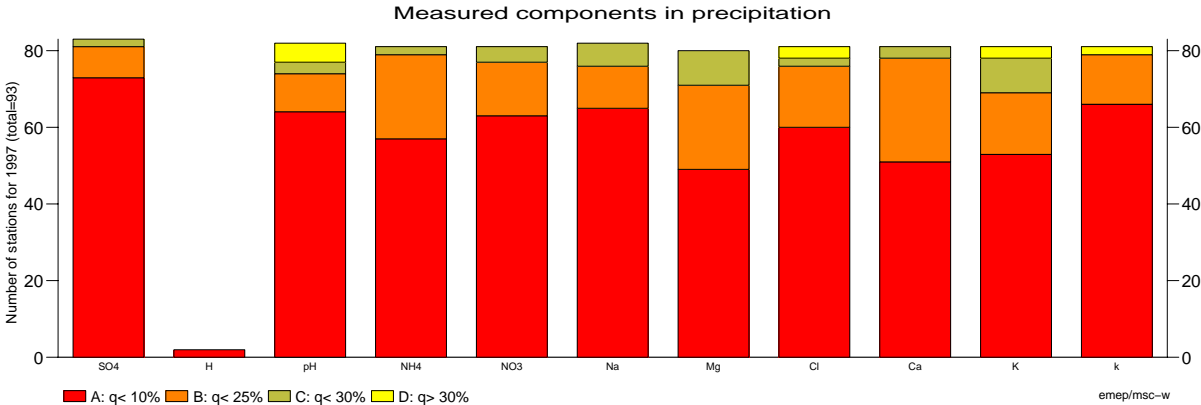
The precipitation program for acidification was followed at the majority of the EMEP stations operative in 1997. Except for H, which was only measured at 2 EMEP sites, 87% of the EMEP stations measure the other required components in precipitation. Measurements are expected to have sampling errors below 25%, for over 83% of the stations despite the fact that wet-only samplers are still not used at the majority of the stations.

- ***On the other hand, sampling sulphur in air needs further harmonisation.***

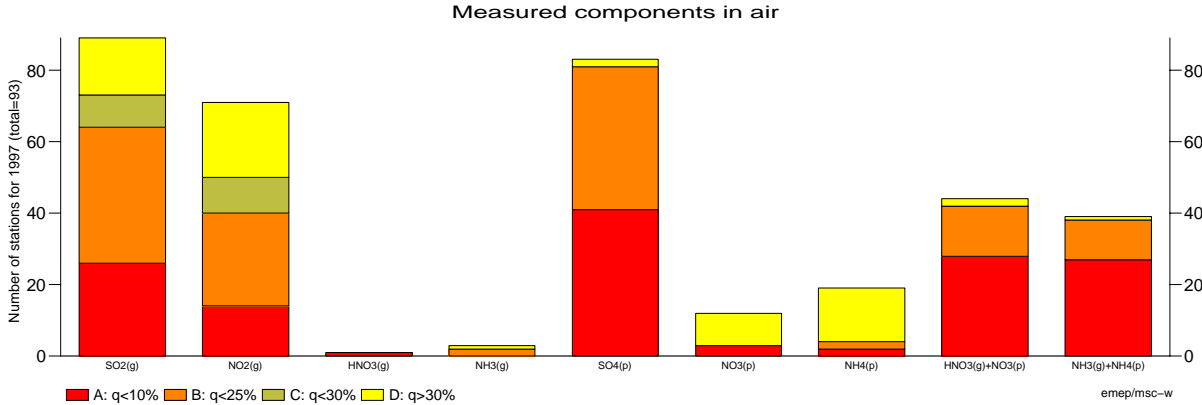
Sulphur dioxide and particulate sulphate are measured at a number (87%) of EMEP stations. The sampling and analysis quality of particulate sulphate measurements is generally satisfactory. For sulphur dioxide, however, there is a wide spread in the quality of the measurements. Below 70% of the SO<sub>2</sub> measurements qualified for sampling uncertainties below 25%. In 1997, 16 EMEP stations were still using non-recommended Thorin or UV fluorescence methods. The UV results are hardly useful for EMEP, and the Thorin measurements have most often a too low quality.

- ***Only 40% of EMEP stations measured nitrogen in air in 1997.***

About 60% of the EMEP stations did not measure all nitrogen components in air in 1997. Only one station, IT1, Montelibretti, reported measurements of gaseous nitric acid. Particulate nitrate has only been measured at three EMEP stations by using the recommended denuder method. Except for nitrogen dioxide measurements, the quality of filter measurements of nitrogen components in air is mostly satisfactory when measured. For nitrogen dioxide, however, there is a wide range of measurement methods in use with variable quality. In particular, monitor data are not useful at regional sites. also, the Salzman method deviates strongly from the recommended method and needs to be replaced.



**Figure 3.2.** Classification of the 1997 precipitation data measurements at EMEP stations. In 1997, the total number of operative EMEP stations was 93.



**Figure 3.3.** Classification of the 1997 air data measurements at EMEP stations. In 1997, the total number of operative EMEP stations was 93.

The quality of EMEP monitoring data has only been studied for 1997. It would be desirable to extend a similar evaluation to the full time series of acidification and eutrophication measurements.

### ***3.4 Suggestions for improvements***

The present EMEP monitoring system could be sufficient to fulfil the main goals of programme, if it was fully implemented to its aims. The 1997 review has shown that the present status of the EMEP network is mostly satisfactory with respect to quality for precipitation data but not for the air data programme. The EMEP centres recommendations and suggestions for future improvements can be summarised as follows:

#### *1) Urgent need for harmonisation: use recommended techniques for sampling and analysis*

The EMEP monitoring system should be strengthened by adopting the same techniques for sampling and analysis in all countries. This is particularly important for future trend analyses and for determination of compliance. QA procedures should be improved and data quality objectives must be adopted at all working levels. Special attention should be given to the harmonisation of sampling and analysis methods for air components.

#### *2) Need for wet-only samplers, it can be discussed whether daily or weekly, but for air programme, daily is absolutely necessary.*

#### *3) Strengthen the measurements of air data in EMEP, in particular, national networks should give priority to the measurements of nitrogen components.*

Only one station (IT1) had a complete air data measurement programme operative in 1997 and determined all seven air components individually including the use of denuders. Only about 40% of the stations used filters to collect the sums of nitric acid/nitrates, and the sums of ammonia/ammonium. In 1997, ~60% of the EMEP stations did not collect all required nitrogen components in air. Monitoring of nitrogen compounds is increasingly important for development and surveillance of the multi-pollutant multi-effect emission control strategies. Nitrogen compounds not only contribute to the acidification and eutrophication of ecosystems but are precursors of tropospheric ozone and they contribute to the total particulate matter.

#### *4) The use of denuders is highly recommended, as it allows the separation of particle and gas components.*

Independent measurements of gas and particle phases are an important requirement for validation of the EMEP assessment. The partitioning between phases affects the transport patterns of the pollutants and thus the influence of transboundary fluxes. This type of measurements are not only important to secure the accuracy of the results from the acidification and eutrophication but are also relevant for the study of atmospheric particulates.

#### *5) Geographical extension to the East and the Mediterranean*

At present there are no EMEP monitoring stations in Armenia, Belarus, Bosnia and Herzegovina, Bulgaria, Cyprus, Georgia, The FRY of Macedonia, the Republic of Moldova, Romania and Ukraine. It would be desirable to extend the EMEP network with at least one station in these countries. There are no operative EMEP stations in Belgium, Luxembourg, Liechtenstein and Malta either. The representativeness of eventual new stations should be assessed.

*6) Enhance country participation (assessment of representativeness)*

The national participation in interpretation and assessment should be enhanced. This refers particularly the question of site representativeness in relation to a larger area, and also to the use of data provided by EMEP in relation to national networks and assessments of concentration and deposition fields at the national level.

*7) Extension of the monitoring with information from other networks*

EMEP should evaluate the possibility of including information from other existing network, in particular within the Convention. For that purpose, it would be desirable to establish closer co-operation with the Working Group on Effects and the ICPs concerning the collection and analysis of measured data.

## *The Operative Eulerian Model*



## ***4. The operative EMEP Eulerian acid deposition model***

For the first time in 1999, the EMEP Eulerian acid deposition model has been used to produce updated estimates of air concentrations and depositions of sulphur and nitrogen and to quantify the significance of transboundary transport over Europe. The introduction of the operational EMEP Eulerian acid deposition model constitutes an important step forward to attaining the operational goals of EMEP on its seventh phase.

This chapter summarises the achievements of the new operative model for acidification and eutrophication, considers its limitations and provides recommendations for further model development. In general, the accuracy of the model results has improved with the present description of physical and chemical processes and the new meteorological input data and the model provides an increased resolution for the calculation of exceedances of critical loads.

Results presented in Bartnicki (1999) show that the new EMEP model is capable of allocating the origin of transboundary air pollution in Europe and manages to account for the “indeterminate deposition” of the previous Lagrangian budget calculations. In source areas, depositions calculated with the new Eulerian model are generally larger than those calculated with the Lagrangian model, but differences between the models are in the ranges of the meteorological variability.

### **4.1 Main objectives for EMEP acid deposition modelling**

The goals of EMEP determine to a great extent the type of model to be used in the programme and impose requirements to the model performance. The EMEP models should provide a scientifically sound description of the physical and chemical processes involved in the atmospheric emission, transport and removal of pollutants. Results from the model should be:

- \* accurate,
- \* mass conservative,
- \* based on state-of-art understanding of physical and chemical processes,
- \* given in appropriate spatial and temporal resolution, and
- \* derived in a computationally effective manner

The accuracy of the model results is measured in terms of their agreement with observations and the validity of the results can also be evaluated through model intercomparisons, sensitivity analysis and sampling error studies. To fulfil these goals operational objectives have been determined inside EMEP.

## 4.2. Present status

After several years of development, the EMEP Eulerian acid deposition model has become ready for operational use. The scientific progress and parametrisations used in the model have been documented and verified since the beginning of its development in 1993 (Berge, 1993; Jakobsen *et al.*, 1995; Jonson and Berge, 1995; Jakobsen *et al.*, 1996; Jakobsen *et al.*, 1997; Berge, 1997; Berge and Jakobsen, 1998, Jonson *et al.*, 1998a, 1998b, Olendrzynski *et al.*, 1998a, 1998b). The model is now operative because it fulfils the EMEP requirements mentioned above.

- 1) On the average, the model performs comparably or better than the EMEP Lagrangian model with regard to measured values. The evaluation of operational model performance is reported in detail in Olendrzynski (1999).
- 2) High precision of mass conservation makes the Eulerian model suitable for the analysis of transboundary exchange and country budgets. The mass conservation properties of the model have been reviewed and improved, leading to pollutant mass conservation within 1% error for the long-term (annual) computations required by EMEP.
- 3) The increased horizontal resolution makes the Eulerian model a better tool for applications on regional scale and for the determination of effects.
- 4) The model has been evaluated and tested to calculate transboundary exchange and source-receptor matrices within reasonable certainty ranges (Bartnicki, 1999)
- 5) The model has been parallelised and optimised for runs in CRAY T3E. The calculations to derive source-receptor matrices for sulphur and nitrogen require 11 CPU hours for each considered country/emitter region.

In addition, a new dedicated meteorological model, PARLAM-PS, has been developed and verified at EMEP/MSC-W. This development has allowed the extension of the EMEP model domain. The new model domain includes now the Turkey and Armenia, both Parties to the CLRTAP Convention that were not totally included in the former domain and allows for a complete description of the Mediterranean Sea.

A detailed description of the meteorological model and its verification for EMEP purposes is given in Tsyro (1999). The need for a new meteorological model is related to the change of EMEP operational transport model from a one-layer Lagrangian model to 3-D Eulerian model, which imposes stronger numerical and physical requirements to the input data. To increase the accuracy of the simulations, input meteorological data is now provided at 3-hour intervals, instead of 6-hourly and the meteorological fields are defined in the same grid as the atmospheric dispersion model to avoid numerical interpolation errors.

The new meteorological model is based on the operational Numerical Weather Prediction at the Norwegian Meteorological Institute (HIRLAM). The HIRLAM model has been

reformulated from a rotated spherical horizontal grid to the polar-stereographic projection, and from a hybrid  $\eta$ -coordinate to  $\sigma$ -coordinate used by EMEP Eulerian models. The usage in both models of the same coordinate system has the great advantage that it avoids mass conservation errors due to spatial interpolation of the meteorological mass and wind fields.

The comparison of HIRLAM/EUROLAM with the meteorological model, LAM50E, used as input for the Lagrangian model has revealed a number of differences in predicted meteorological fields which can be relevant to explain differences in concentrations and deposition fields computed for 1997. The largest differences are found in cloud cover, precipitation, and surface fluxes of heat and momentum. An interesting difference between the meteorological models is the parametrisation of cloud water and the explicit treatment of evaporation below cloud in the HIRLAM. This introduces new possibilities for the wet scavenging parametrisation of the Eulerian model which are worth investigating in the future.

#### **4.2.1. Evaluation of Eulerian model performance**

The accuracy of model results is usually determined by their agreement with observations and for EMEP purposes, the operative quality standard for yearly averages has been “agreement within a factor of two”.

The evaluation of operational model performance reported in Olendrzynski (1999) focuses on yearly and daily values. Scatter diagrams demonstrate that the computed annual mean air concentrations are generally within a factor of two agreement with observations in most of the EMEP stations in 1997. For  $\text{SO}_2$ , however, the model tends to overestimate high concentrations close to emission sources, and to underestimate low concentrations in remote areas of Europe.

It has been pointed out before that the dry deposition routine of the Eulerian model does not perform satisfactorily (Jakobsen et al., 1996). In Olendrzynski (1999) a test with fixed dry deposition velocity at 1m for  $\text{SO}_2$ , showed that the agreement with measurements improves significantly for  $\text{SO}_2$ , while the results for other components are - except  $\text{SO}_4$  - only slightly affected. This is again an indication of the need to revise the computation of surface and quasi-laminar boundary layer resistances in the model. A closer look into this problem is intended in the near future.

For compounds other than  $\text{SO}_2$ , computed annual mean air concentrations are in relatively good agreement with observations. The difference between computed and measured air concentrations, averaged over all stations considered, varies between 1-13% with  $\text{NO}_2$  and  $\text{NH}_3+\text{NH}_4$  having the best agreement. Correlation coefficient between computed and measured concentrations is rather high, varying between 0.69 and 0.87 depending on the compound. Computed annual concentrations in precipitation are also in fairly good agreement with observations, where between 74-87% of all stations are within the zone of ‘the factor of two’ difference. Table 4.1 summarises the performance of the Eulerian model with respect to yearly averages and compares it with the averaged performance of the Lagrangian model.

Analysis of the daily measured and computed concentrations indicated that the dynamics of the concentration changes is frequently well captured by the model. This is particularly evident at stations in Scandinavia on occasions of long-range transport from continental Europe.

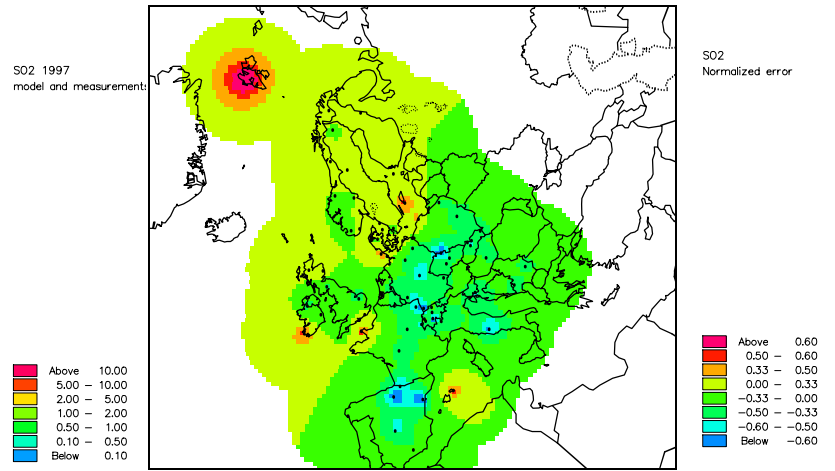
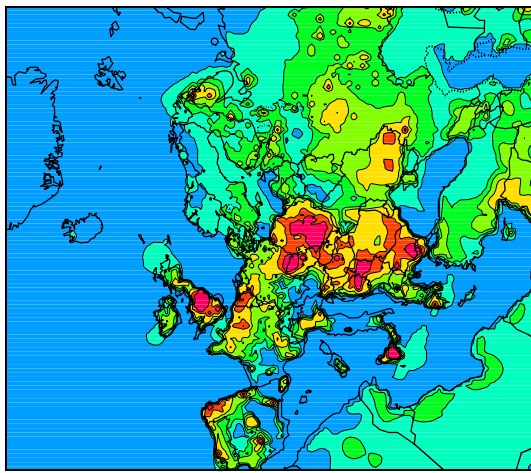
Model performance for southern Europe is, in general less satisfactory than for the rest of the continent.

**Table 4.1.** Comparison of the performance of the Eulerian and the Lagrangian models with respect to yearly averaged concentrations.

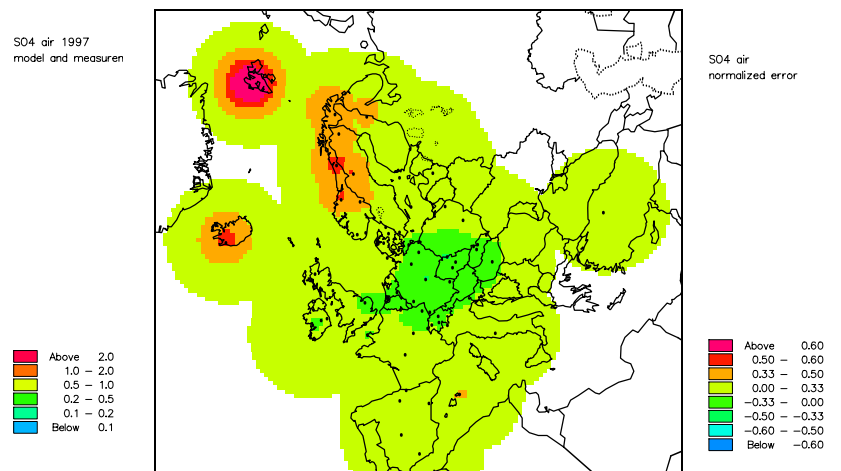
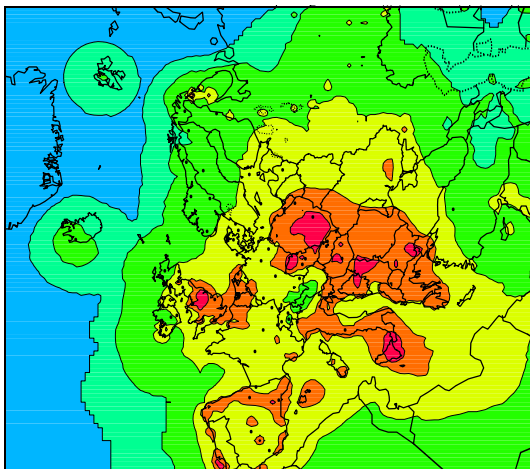
Component	Eulerian 1997				Lagrangian 1985-97			
	daily correl	Mod Mean	Obs Mean	Rel. Bias	year. correl	Mod Mean	Obs Mean	Rel. Bias
SO <sub>2</sub>	0.74	2.88	1.51	90%	0.87	2.73	2.81	3%
SO <sub>4</sub>	0.84	0.73	0.83	12%	0.79	1.57	1.27	3%
NO <sub>2</sub>	0.80	2.06	2.04	1%	0.82	1.25	1.91	35%
HNO <sub>3</sub> +NO <sub>3</sub>	0.69	0.62	0.55	13%	0.96	0.86	0.32	168%
NH <sub>3</sub> + NH <sub>4</sub> <sup>+</sup>	0.53	1.25	1.36	8%	0.93	0.56	0.78	28%
SO <sub>4</sub> <sup>-2</sup> (l)	0.64	0.72	0.51	41%	0.67	0.78	0.82	5%
NO <sub>3</sub> <sup>-</sup> (l)	0.59	0.36	0.35	3%	0.68	0.44	0.45	2%
NH <sub>4</sub> <sup>+</sup> (l)	0.64	0.48	0.42	14%	0.83	0.37	0.58	36%

Following the approach introduced in Tarrasón et al. (1998), combined estimates of the geographical distribution of sulphur and nitrogen compounds have been derived. A similar approach has also been followed in Kåresen and Hirst (1999) for sulphur components, where in addition, confidence ranges of the estimates are presented.

The general agreement between the Eulerian model results for 1997 and EMEP measurements is better than for the Lagrangian model, specially for nitrogen dioxide and nitrate in air. Figures 4.1 and 4.2 show the results for sulphur dioxide and particulate sulphate in air, together with the normalised differences between the Eulerian model estimates and 1997 EMEP measurements. These are the two components with worse model performance. The systematic overestimate of particulate sulphate in central Europe and underestimation in Scandinavia has been related before to the  $\beta$ -factor, the fraction of emissions directly emitted as particles. The combination of model and measurement estimates are the present best estimate for particulate sulphate in air and sulphur dioxide.



a)



b)

**Figure 4.1.** Combined estimate of modelled and measured data and normalised differences for for a) sulphur dioxide and b) particulate sulphate in air . Units:  $\mu\text{g}(\text{S})/\text{m}^2$

#### 4.2.2. Evaluation of the Eulerian source-receptor matrices

Non-linear chemistry and non-linear effects induced by the numerical solution of the transport equation are the major problems in computing source-receptor matrices with the Eulerian Acid Deposition model. In practice, these non-linearities imply that sum of individual contributions is not necessary equal to the total deposition from all European sources and the differences between the total and separate runs indicate the range of uncertainties in the source-receptor matrices due to non-linear effects.

Bartnicki (1999) presents a thorough evaluation of different methods to calculate source-receptor matrices with the new Eulerian model and quantifies the ranges of uncertainties in the annual source-receptor matrices due to non-linear effects to be 4% for oxidised sulphur 8% for oxidised nitrogen and 3% for reduced nitrogen. The actual source-receptor matrices are also given in Chapter 1.3.

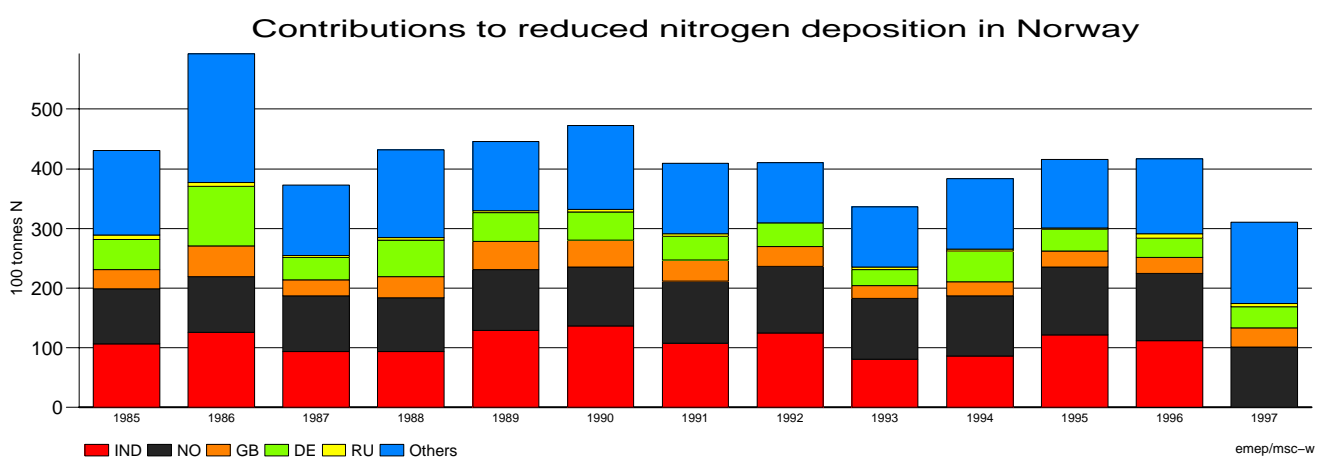
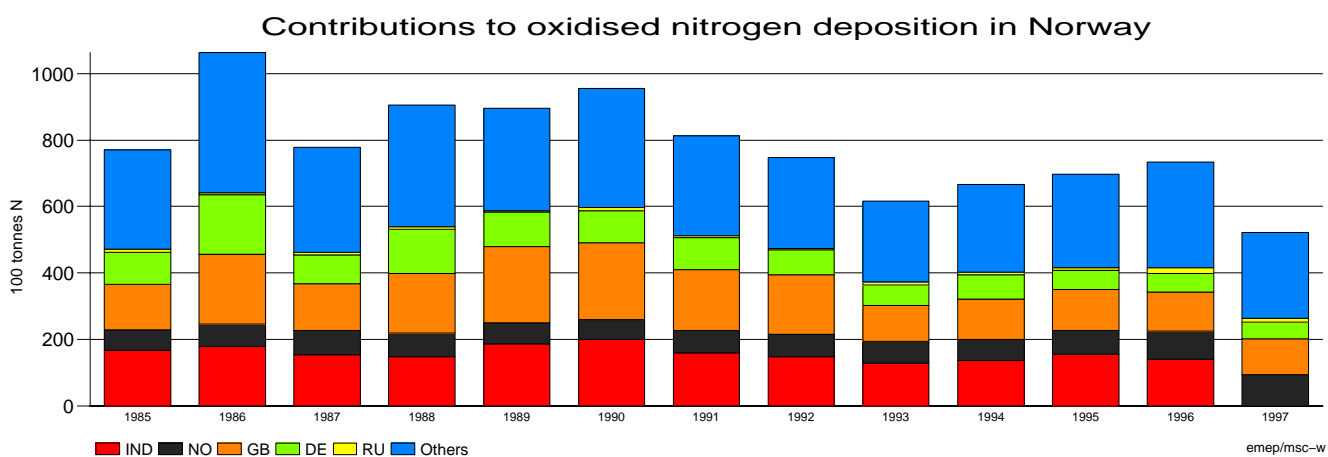
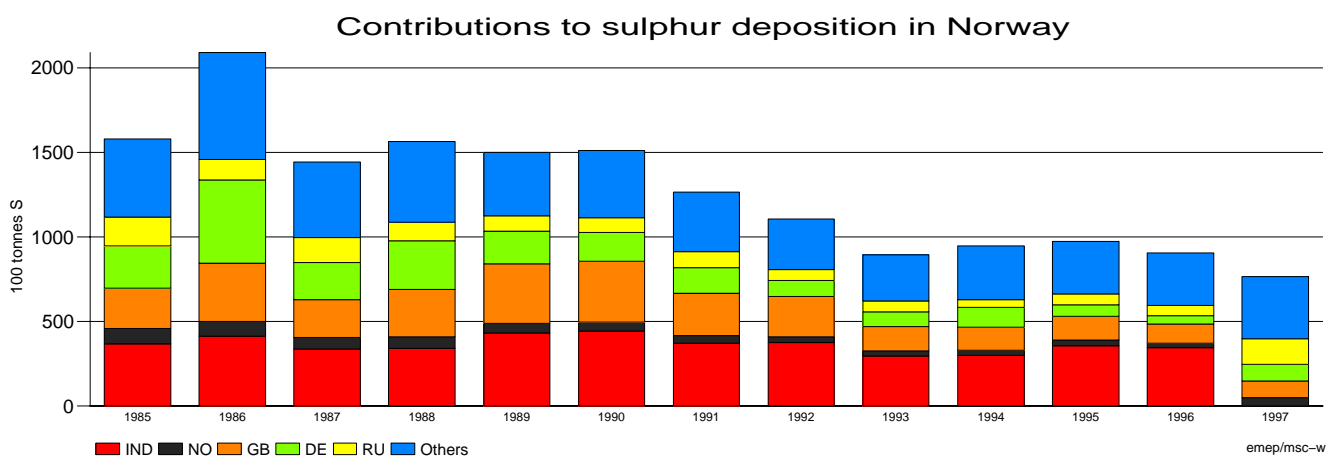
Given the consequences for the work of the Convention, Bartnicki (1999) has compared the Eulerian model matrices with the source-receptor matrices computed with the Lagrangian model. Both models gave similar results when the input meteorological data were computed by the LAM50E meteorological model.

In general, the Eulerian model allocates a larger fraction of the European emissions and the manages to account for the “indeterminate deposition” of the previous Lagrangian budget calculations. This is well illustrated in Figures 4.2, 4.3 and 4.4 which show the evolution of the five larger contributors to deposition over Norway, Spain and Poland.

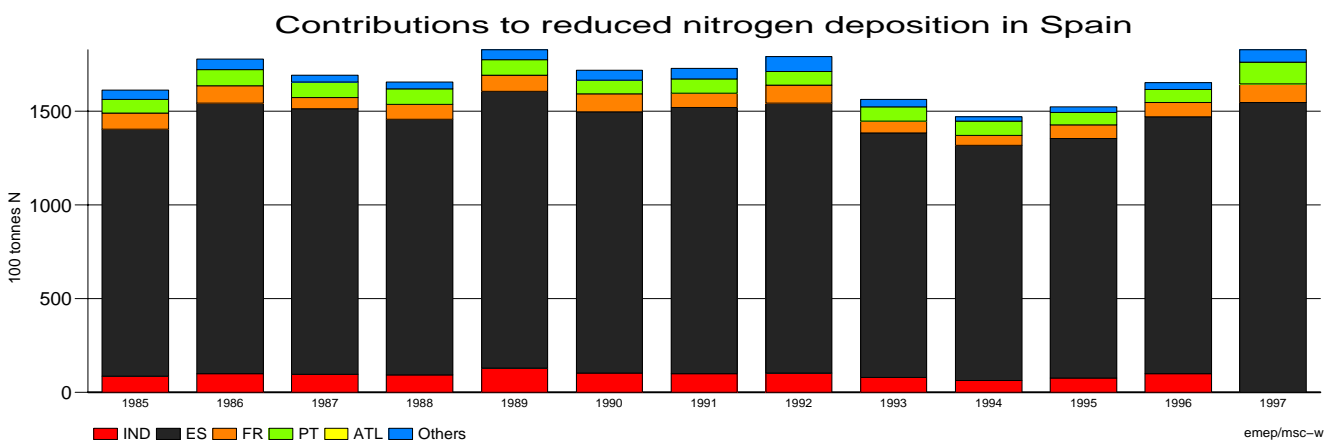
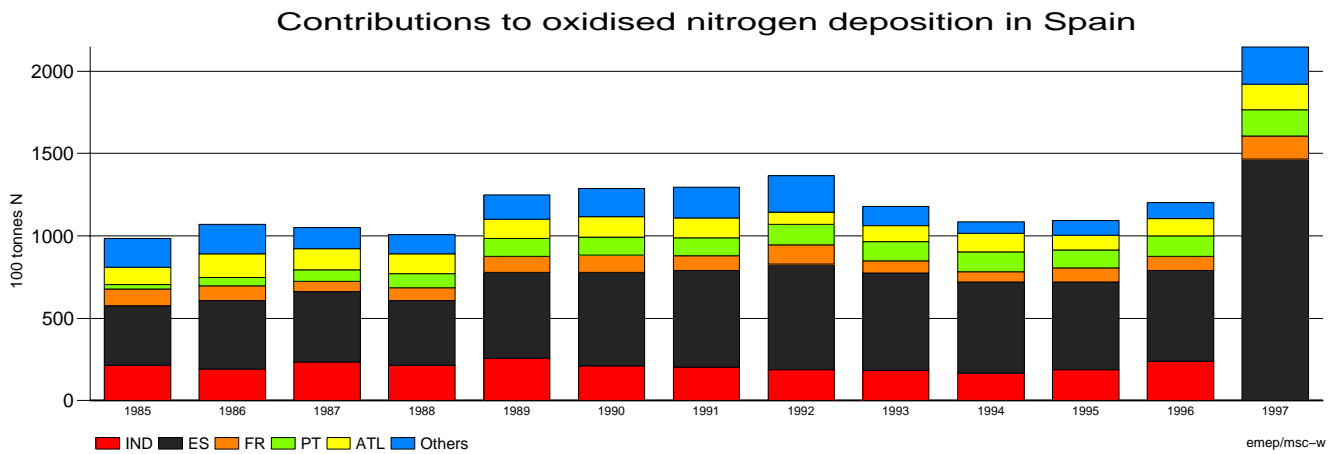
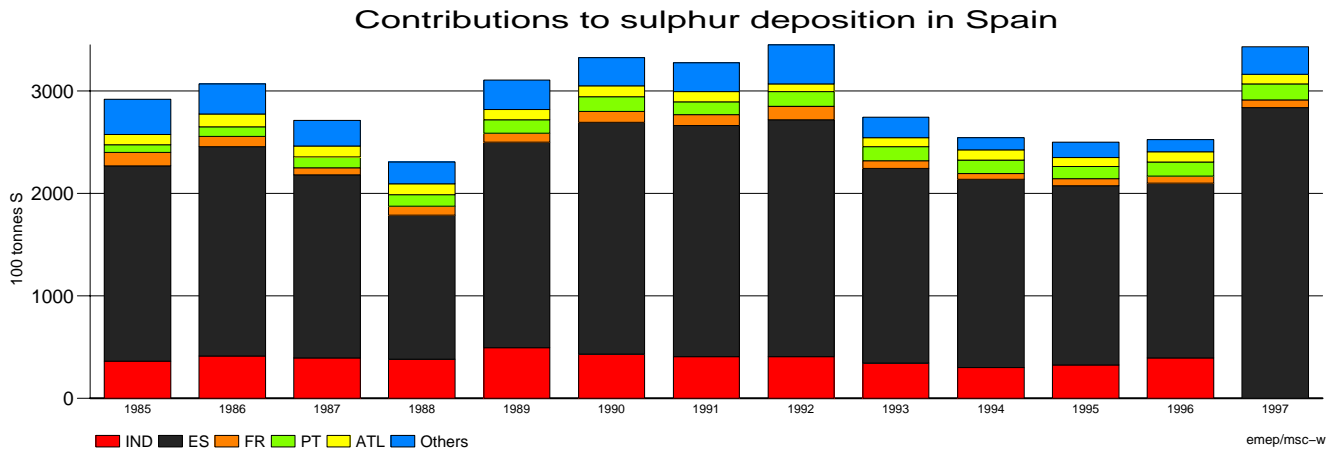
The Eulerian model manages to allocate the “indeterminate” contribution thus increasing the actual contribution of different countries. In many cases, the contribution of the country to itself increases in the 1997 simulations. This is particularly true in source areas and for Mediterranean countries. The deposition of Spanish emissions in Spain increases in 1997 with respect to previous years, particularly for oxidised nitrogen and this seems to be related to an increase in the dry deposition calculated by the Eulerian model.

It is not clear at this point whether this is a feature of the Eulerian model or if it is a consequence of the particular meteorological variations of this year. The generally larger contribution of dry deposition to total in 1997 can be to some extent explained by the differences in precipitation, which was approximately 20% lower in 1997 than in 1996 in the input files for the Eulerian model. Further studies using different meteorological conditions are necessary.

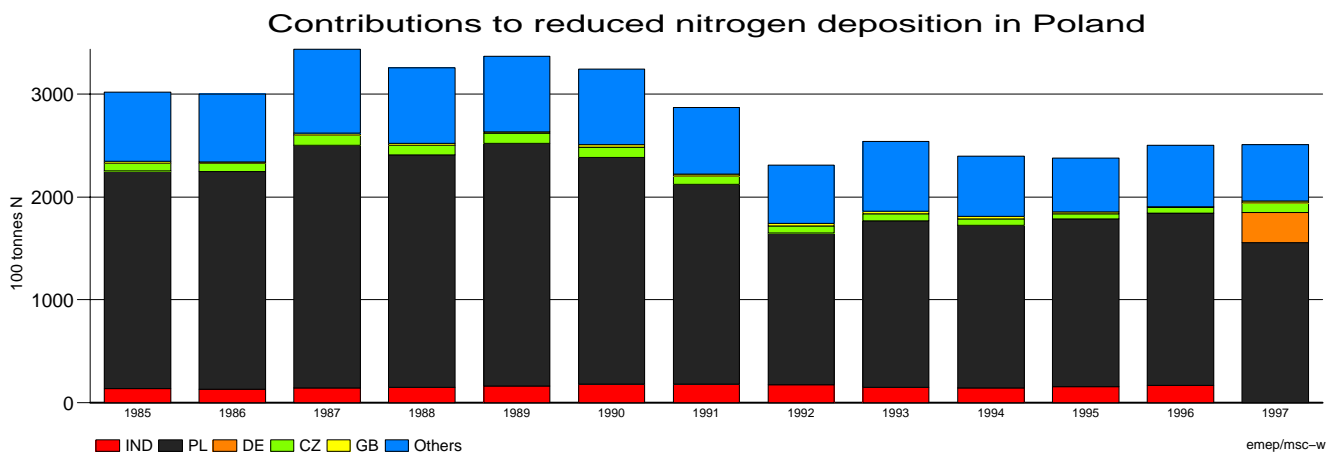
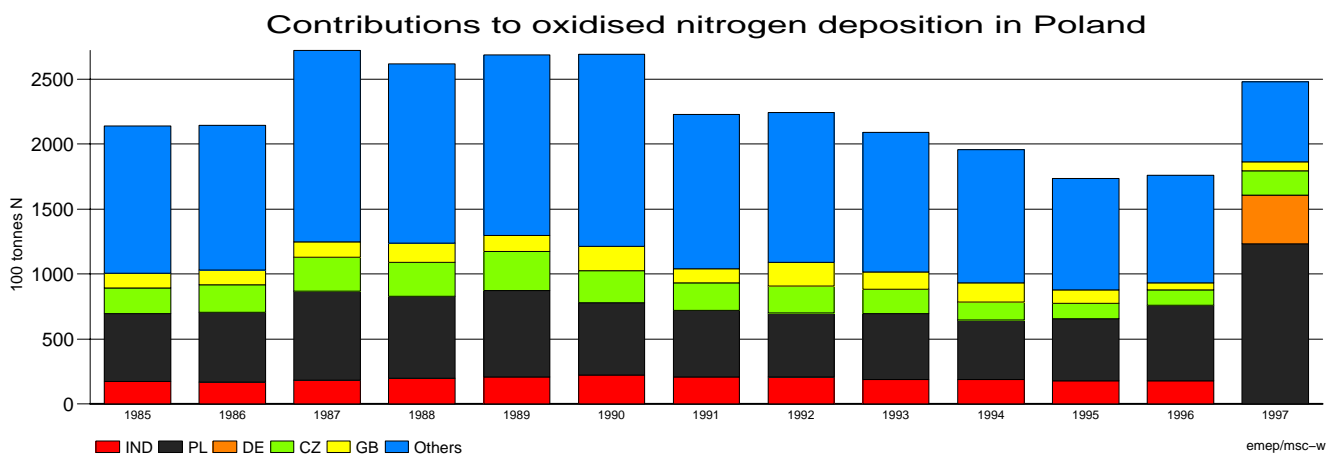
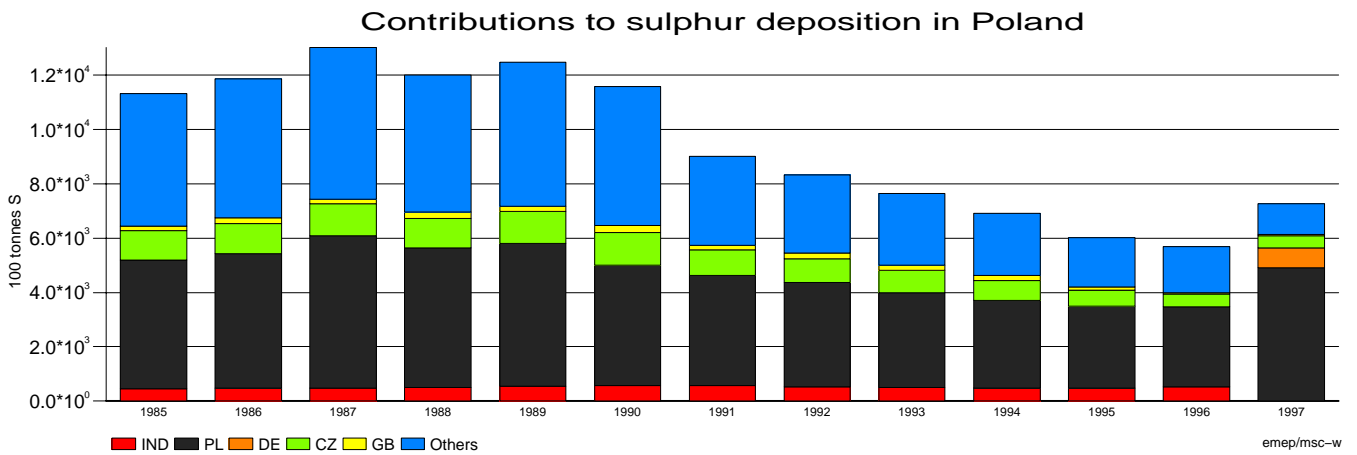
Otherwise, differences between the source-receptor matrices produced by Eulerian and the Lagrangian models are within the range expected from different meteorological variations.



**Figure 4.2.** Evolution of allocated deposition over Norway. Five main contributors as reported in the Lagrangian source-receptor matrices for the period 1985-1996, and the Eulerian source-receptor matrices in 1997.



**Figure 4.3.** Evolution of allocated deposition over Spain. Five main contributors as reported in the Lagrangian source-receptor matrices for the period 1985-1996, and the Eulerian source-receptor matrices in 1997.



**Figure 4.4.** Evolution of allocated deposition over Poland. Five main contributors as reported in the Lagrangian source-receptor matrices for the period 1985-1996, and the Eulerian source-receptor matrices in 1997.

### **4.2.3. Statistical estimates of emissions using of source-receptor matrices**

Previous studies reported by the MSC-W concluded that the use of statistical methods to determine the level of compliance to the protocols of the Convention protocols requires a qualification of the uncertainties associated both with model and measured concentrations in the EMEP programme (Dimakos and Høst, 1997). Their original analysis was inconclusive because it assumed that all deviations between EMEP modelled and measured data was caused by inaccuracies in the emissions. By not accounting for model uncertainties, they reached conflicting results on the extent of the emissions depending on the atmospheric sulphur component that was analysed.

This year, Kåresen and Hirst (1999) report further on the use of a Bayesian method to evaluate emissions in the EMEP area. Following the recommendations from previous studies they include an estimate of the measurement error in the analysis. The measurement error that they have used was derived by Aas et al. (1999) and corresponds to the quality analysis reported in Chapter 3.

They conclude that inclusion of the given uncertainty ranges in the measured data has little effect on the results. Their estimated changes in the emissions are not significant and they are uncorrelated between the different sulphur components, as in Dimakos and Høst (1997). In this way, they conclude that there is insufficient information in the EMEP data to evaluate national emissions. This could have consequences for the geographical distribution of EMEP stations, but given the uncertainties in this model approach it is not recommended to use statistical Bayes estimates based on the EMEP source-receptor matrices to evaluate emission inventories.

## 4.4 Recommendations for future model development

The introduction of the operational EMEP Eulerian represents a first step towards the development of combined multi-pollutant, multi-effect models. Its further development should be parallel to the development of the Eulerian photooxidant model. The models having the same structure, it is important that new routines are tested and implemented in both models. The EMEP Eulerian model is envisaged to evolve towards a flexible model useful for a wide range of policy relevant applications.

An important goal for future model development should be to improve the accuracy of model results to at least a 30% agreement with measurements for all components. This implies, among other things, the need to:

### *1) Review the dry deposition parametrisation*

Sulphur dioxide in air shows poor agreement with observations even when annual averages are considered. The model tends to overestimate high concentrations close to emission sources, and to underestimate low concentrations observed in remote areas of Europe. Tests performed this year with dry deposition indicate that the formulation of the surface and the sub-laminar resistances can be a source of error in the model. It is expected that an improvement on the dry deposition routine will lead to an increase in the accuracy of the model estimates for sulphur dioxide in air.

### *2) Review wet scavenging parametrisation to account for re-evaporation.*

Sulphate in precipitation is the other component where the EMEP Eulerian model does not satisfy yet the accuracy requirement of 30% agreement in the yearly means, and is generally overestimated by the model. This recommends a review of the wet scavenging routine in order to introduce the effect of re-evaporation processes. The possibility of including re-evaporation in the Eulerian model formulation is provided by the introduction of the new HIRLAM\_PS. However, the hydrological balance in the new meteorological model should be evaluated during the study.

### *3) Improve the chemical scheme by comparing with the parametrisation of the Eulerian photo-oxidant model.*

In particular the sulphur chemistry and the partitioning between  $\text{HNO}_3(\text{g})$ - $\text{NO}_3(\text{p})$ , processes that have further importance further in modelling atmospheric secondary aerosols.

### *4) Explore new methods for evaluation of model performance, that consider the challenges*

### *5) Investigate new inverse methods to report on compliance of emissions.*



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***APPENDIX***  
***1997 Status Review***  
***by Country***

## Overview of regions included in this Appendix

Country/Region	Code	Country/Region	Code
Armenia	AM	Norway	NO
Austria	AT	Poland	PL
Belarus	BY	Portugal	PT
Belgium	BE	Republic of Moldova	MD
Bosnia and Herzegovina	BA	Romania	RO
Bulgaria	BG	Russian Federation	RO
Croatia	HR	Slovakia	SK
Cyprus	CY	Slovenia	SI
Czech Republic	CZ	Spain	ES
Denmark	DE	Sweden	SE
European Union	EU	Switzerland	CH
Finland	FI	The FYR of Macedonia	MK
France	FR	Turkey	TR
Germany	DE	Ukraine	UA
Greece	GR	Yugoslavia	YU
Hungary	HU	United Kingdom	GB
Iceland	IS		
Ireland	IE	Estonia	ES
Italy	IT		
Latvia	LV	The Baltic Sea	BAS
Lithuania	LT	The Mediterranean Sea	MED
Luxembourg	LU	The North Sea	NOS
Netherlands	NL	Remaining N.E. Atlantic	ATL

**For Canada, Liechtenstein, United States only reported emission data**

**Georgia and Malta not included at present.**



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