

Air Pollutants in Germany: Long Term Trends in Deposition and Air Concentration

Th. Gauger & F. Anshelm

Institute of Navigation, Stuttgart University, Geschwister-Scholl-Str. 24 D,
D-70174 Stuttgart, Germany

Abstract

Within the German Critical Levels and Critical Loads mapping activities, led and funded by the German Federal Environmental Agency (UBA), mapping of deposition and air concentration of several relevant air pollutants as well as related effects on ecosystems and materials are carried out. The mapping approaches are using the commonly agreed procedures of the international co-operative programme for mapping Critical Loads and Levels and geographical areas where they are exceeded, which is guided by the ICP on Modelling and Mapping. The German mapping monitors the long-term trends of air pollutants and their effects on a national scale by making use of all available data of the deposition and air concentration monitoring networks in Germany.

Since the start of the ICP Materials material exposure programme in 1987 considerable changes concerning geographic distribution and range or amount of air pollutants over Europe and Germany can be observed. German maps and statistics showing trends in air pollution are presented and discussed. Due to the political and economical changes that started at the end of the 1980ies in Eastern Europe and the ongoing success of emission abatement, a 77% reduction of in air sulphur dioxide (SO_2) can be observed between 1987 and 1995. Wet deposition loads of sulphur ($\text{SO}_4\text{-S}$) and chloride, corrected for sea salt contribution ($\text{Cl}_{(\text{ssc})}$), remarkably diminished by about 53% and 71% respectively (Table 1). No clear trend of reduction, however, can yet be observed in the annual average air concentration levels of nitrogen dioxide (NO_2) and ozone (O_3) and in the wet deposition rates of protons (H^+).

1 Introduction

Within the German Critical Levels and Loads research and mapping activities on behalf and the account of the German Federal Environmental Agency (UBA), mapping of deposition loads and air concentration of the main air pollutants is carried out at Institute of Navigation, Stuttgart University. The research carried out in the project 'Critical air concentration levels and deposition loads and their exceedance in agricultural, forest and other non-forest semi-natural ecosystems' has yielded, as one product among others, long-term time series of maps of main air pollutants. The preliminary German maps are presented here in order to illustrate the changes in the amount and geographic distribution of air pollutants in time. The differences of annual average wet deposition loads and air concentration of the different species mapped in 1987, 1992 and 1995 respectively, is shown in Table 1.

Table 1: Differences of average wet deposition loads and air concentration levels in Germany

Wet Deposition	1987 eq/ha·a	1995 eq/ha·a	Diff. %	Air Concentration	1987 µg/m³·a	1992 µg/m³·a	1995 µg/m³·a	Diff. %
SO ₄ -S	1041	491	-53%	SO ₂	48	--	11	-77%
SO ₄ -S _(ssc) [*]	1013	462	-54%	NO ₂	33	--	27	-18%
NO ₃ -N	355	295	-17%	NO	--	28	21	-24%
NH ₄ -N	479	401	-16%	NO _(B) ^{**}	--	16	15	-9%
N = (NO ₃ -N + NH ₄ -N)	834	697	-16%	NO _x	--	78	62	-21%
Ca _(ssc) [*]	316	113	-64%	NO _{x(B)} ^{**}	--	55	48	-13%
Mg _(ssc) [*]	46	9	-79%	O _{3(B)} ^{**}	--	45	46	+3%
K _(ssc) [*]	43	29	-33%					
BC _(ssc) [*] = (Ca _(ssc) + K _(ssc) + Mg _(ssc))	404	151	-63%					
Cl	314	272	-13%					
Cl _(ssc) [*]	48	14	-71%					
Na	242	241	0%					
H	402	225	-44%					
* _(ssc) = sea salt corrected (anthropogenous only)				** _(B) = Background (rural measurements only)				

Main database for mapping air concentration and wet deposition are the routine measurements of the air concentration and wet deposition monitoring networks in Germany. The data of the different species, measured at about 110 to more than 400 measurement points (Table 2), are interpolated using kriging technique to derive air concentration and wet deposition fields respectively.

The geographical data sets derived by mapping air pollutants in Germany are used as input for the calculation of Critical Loads and Critical Levels exceedance in ecosystems as well as for mapping actual corrosion rates and the exceedance of acceptable corrosion rates for materials. Results of the latter are presented in the paper of Anshelm et al. 'Mapping actual corrosion rates and exceedances of acceptable corrosion rates - procedure and results' in the workshop proceedings.

2 Air concentration

Air concentration fields are derived by kriging interpolation of air concentration measurement data. The input data are provided by the German Federal Environmental Agency (UBA), Berlin. In the single years due to the installation of measurement sites, the amount of available data differs (Table 2). The number and geographical scatter of the measurement sites, however, is considered appropriate to map the spatial trend of air concentration over Germany.

Table 2: Number of input data for mapping air concentration fields in Germany

Year	SO ₂	O ₃	NO ₂	NO	NO _x
1985	289	-	179	-	-
1986	308	-	193	-	-
1987	380	-	234	-	-
1988	413	-	241	-	-
1989	492	-	219	-	-
1990	405	110	330	-	-
1991	491	134	350	-	-
1992	496	157	377	179	174
1993	503	175	353	203	188
1994	516	186	410	138	132
1995	492	212	433	248	232
1996	475	224	459	294	289
1997	457	230	451	249	246
1998	436	227	448	270	269

2.1 Sulphur dioxide (SO₂)

A fourteen year time series of annual maps is calculated for sulphur dioxide (SO₂), beginning in 1985. Referred to annual average values, SO₂ air concentration reduces 88% between 1985 and 1998 (Figure 1). The reduction of in air SO₂ to a great extent reflects the clear downward trend of emission observed on the European and German scale (Vestreng & Støren 2000). The reasons for the emission reduction can be found in the efforts of emission abatement and the political and economic changes that started at the end of the 80ies in Eastern Germany and in the Eastern European countries.

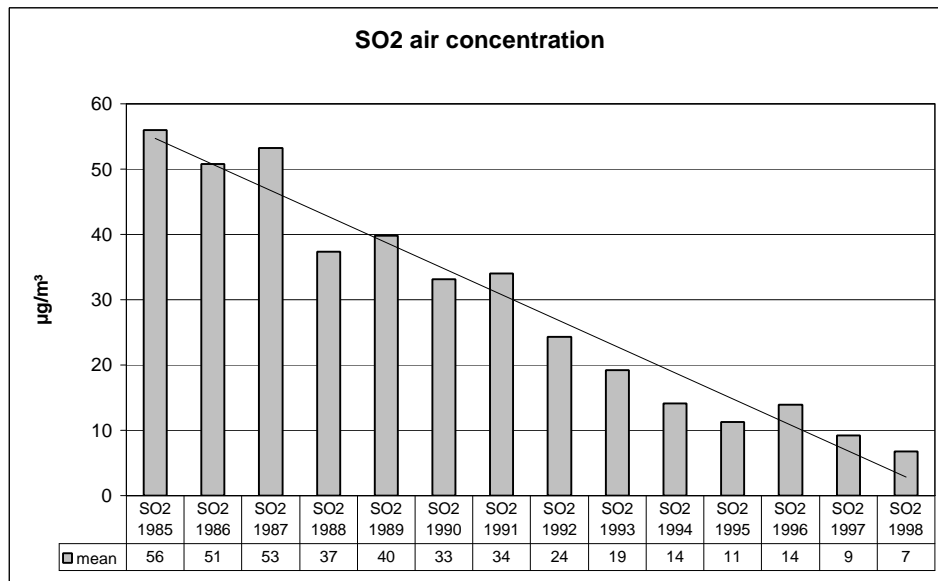


Figure 1: Annual average SO₂ air concentration in Germany 1985-1998

The geographical distribution of in air SO₂ over Germany and its changes are shown in annual maps (Figures 2 - 4). In the first three years 1985 to 1987, high concentration levels do not change very much. From 1987 to 1988 a remarkable reduction of about 30% can be observed. The decline of SO₂ air concentration mainly is found in the western part of Germany, whereas the situation remains more or less unchanged in the former German Democratic Republic (Figure 2).

The whole situation only slightly changes from 1988 until 1991. In the following years 1991 to 1995 (Figure 3), average SO₂ air concentration diminishes continuously about 20% to 30% from one year to the next. Areas of SO₂ air concentration above 30 µg/m³ mainly covering Eastern Germany, more and more disappear from the maps (Figure 3 and 4). From the year 1995 to 1996 (Figure 4) the average SO₂ air concentration on average rises by about 24%. Higher air concentration measurement values mainly are found at the beginning of 1996. Further analysis of data and literature showed that the main reason for higher SO₂ can be found in the relatively cold winter period in early 1996. The average SO₂ air concentration in 1997 again shows a reduction by about 34% compared to 1996. In 1998 the average SO₂ level in Germany is 27% lower than in 1997.

Highest SO₂ air concentration values in all years are found in eastern Germany, namely in Saxony, the German part of the "black triangle" area.

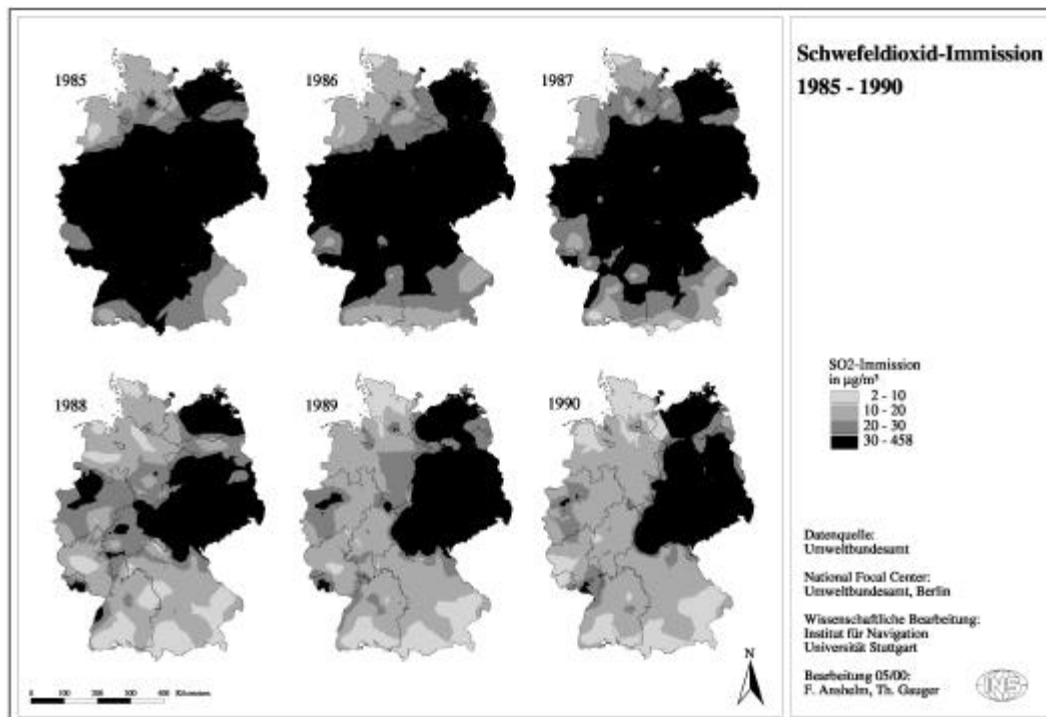


Figure 2: Annual average air concentration of SO₂ in the years 1985 to 1990

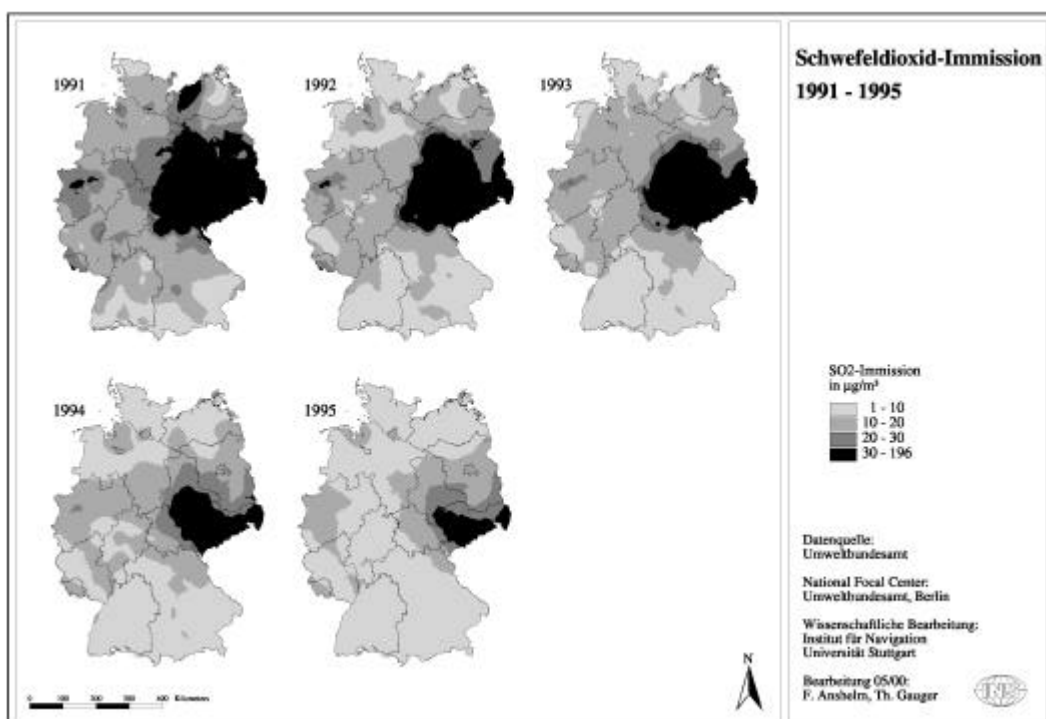


Figure 3: Annual average air concentration of SO₂ in the years 1991 to 1995

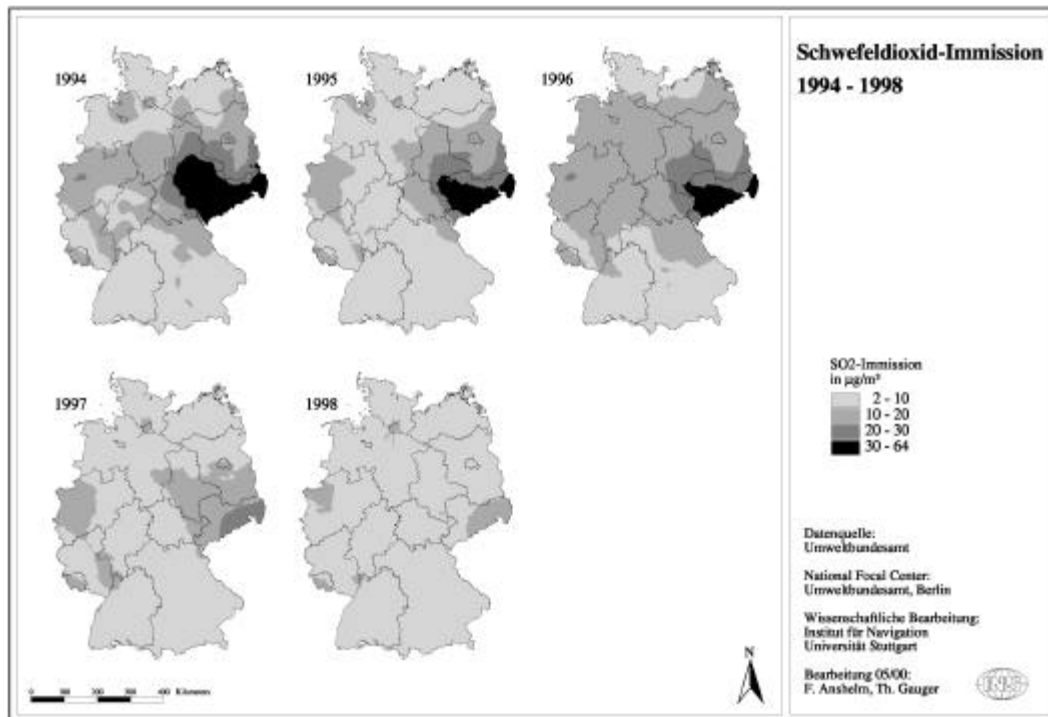


Figure 4: Annual average air concentration of SO₂ in the years 1994 to 1998

2.2 Nitrogen dioxide (NO₂)

In contrast to the observed reduction of in air SO₂, no trend of reduction of nitrogen dioxide (NO₂) air concentration can be found (Figure 5). The range of differences in annual average NO₂ air concentration is +/- 1% to 16%.

In Figure 6 the maps of annual average in air NO₂ in the years 1991 to 1995 are presented. The spatial distribution of NO₂ (Figure 6) is very similar each year: In years of higher average NO₂ values, e.g 1993, the legend class above 40µg/m³ is extended over larger areas. Highest values (close to 70 µg/m³) can be found around some urban/industrial agglomeration areas, whereas mainly rural regions show lower NO₂ air concentration.

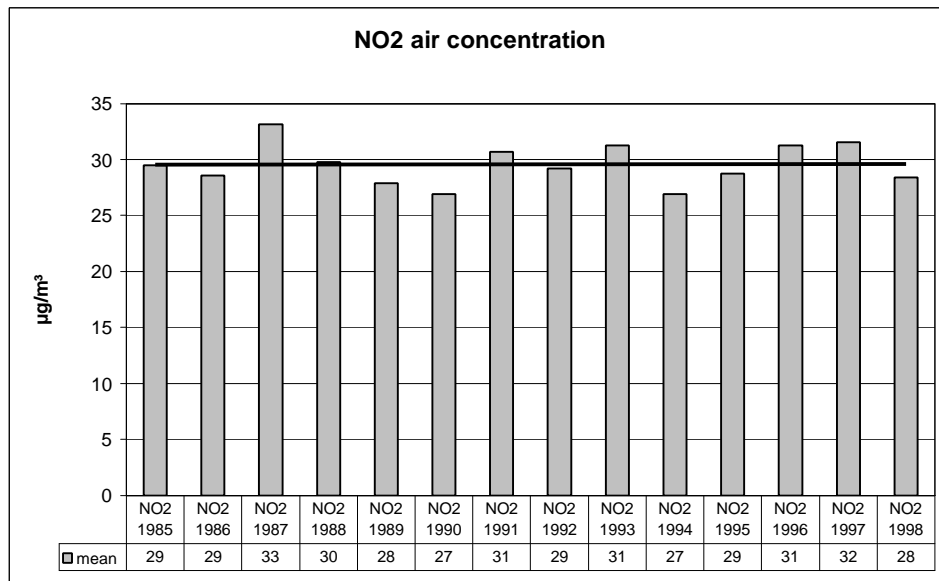


Figure 5: Annual average NO₂ air concentration in Germany 1985-1998

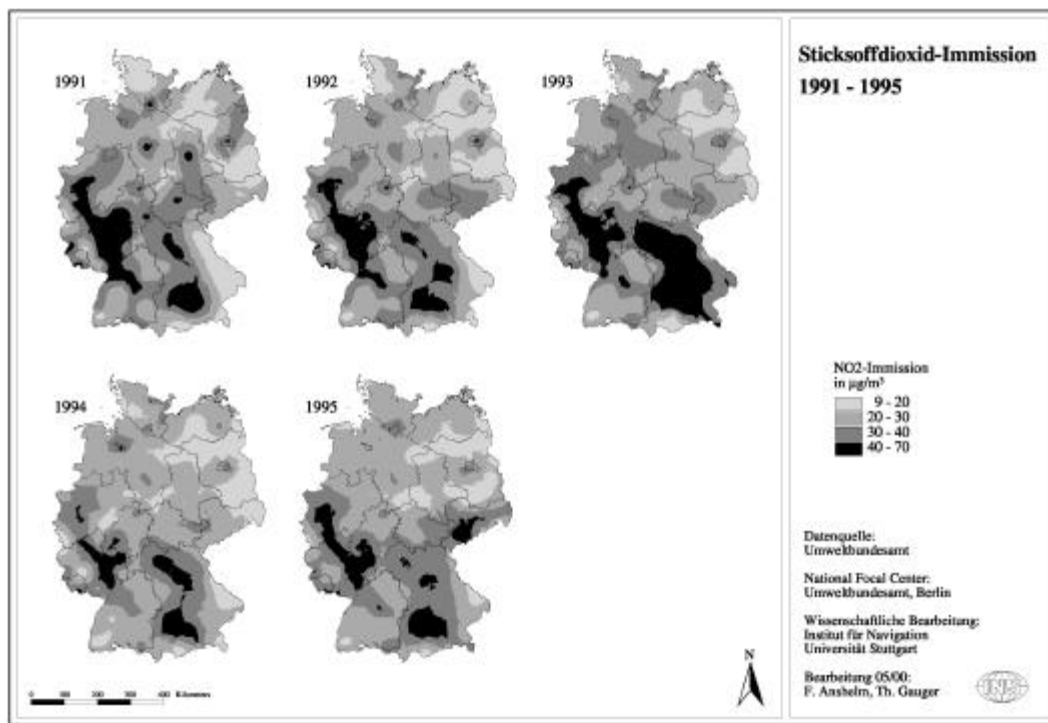


Figure 6: Annual average air concentration of NO₂ in the years 1991 to 1995

2.3 Ozone (O₃)

Ozone maps are calculated for the years 1990 to 1998. In Figure 7 the oscillation of the average values can be observed. The differences between average annual air concentrations range between -5% and +8% and are mainly due to the climatic and summertime weather conditions of the respective year. No general trend can be observed, though the 1998 average is 10% higher than in 1990.

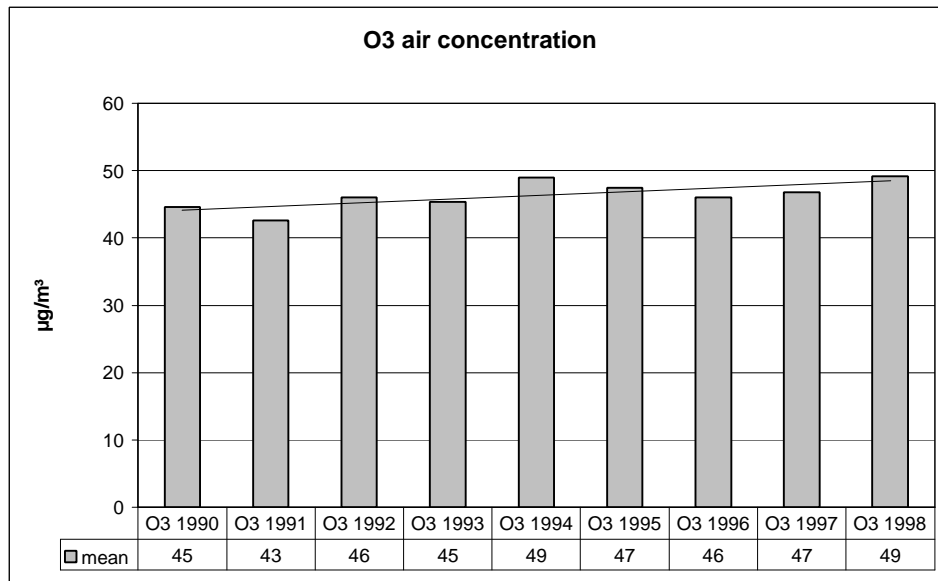


Figure 7: Annual average O₃ air concentration in Germany 1985-1998

In the 1991 to 1995 maps shown in Figure 8 the annual differences of in air O₃ are reflected in the variation of the extent of high and low value areas.

In contrast to the NO₂ maps shown above, highest O₃ values are found in remote rural areas, whereas in and around urban/industrial agglomeration areas low O₃ concentrations are found. This is due to the reaction of ozone with nitric oxide (NO), which rapidly generates nitrogen dioxide (NO₂) in areas of high NO concentration, typical for urban areas with high vehicle exhaust emissions. Maximum values of O₃ are found at remote mountain areas of high elevation, which are less influenced by the depletion of ozone by nitric oxide (Figure 8).

In Figure 8 also the average O₃ concentration over a period of five years (1991-95) is presented. Because of large year to year variations, the annual average of a five year period more clearly shows the occurrence of high and low ozone concentration in certain regions.

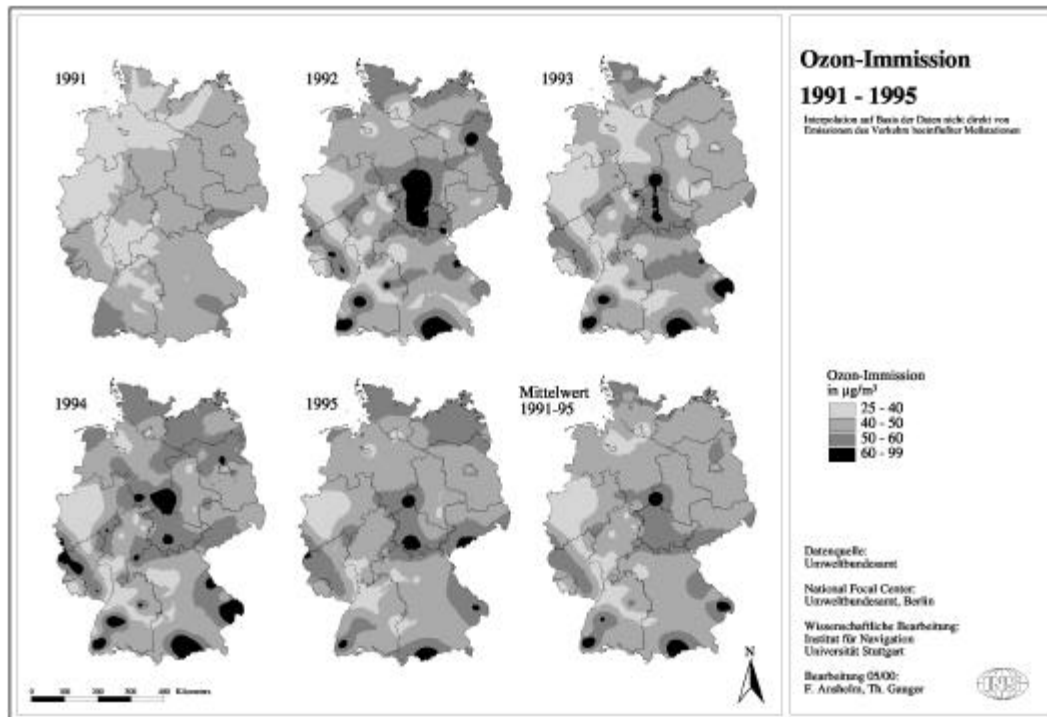


Figure 8: Annual average air concentration of O₃ in the years 1991 to 1995 and average O₃ air concentration in the five year period 1991-95

3. Wet deposition

The mapping of wet deposition loads in Germany as well as the mapping of air concentration fields is based upon monitoring data. A wet deposition database has been set up at Institute of Navigation (INS) by comprehensive data acquisition at the institutions responsible for local, regional and national deposition monitoring programs in Germany. The mapping approach is carried out according to the recommendations of the UN/ECE Mapping Manual (UBA 1996). The outline of the mapping procedure can be seen in Figure 9, which shows the example of mapping 1993 sulphur (SO₄-S) wet deposition. Point measurement data of solute concentration (Figure 9 A) are interpolated using kriging technique to derive solute concentration fields (Figure 9 B). In a next step a sea salt correction is carried out to subtract the sea spray contribution to wet deposition of the base cations (BC) calcium (Ca), magnesium (Mg) and potassium (K) and of chloride (Cl) and sulphur (SO₄-S). Sodium (Na) is assumed to be exclusively of sea salt origin. Na wet deposition is used as tracer for sea salt influence, together with a factor, which is the ratio of sodium and the other components in seawater to calculate non marine wet deposition (SO₄-S_(SSC), Figure 9 C). The map of solute concentration fields (in meq/l) then is multiplied with a precipitation map, provided by the German Meteorological Survey (DWD) (Figure 9 D), to derive maps of wet deposition loads (in eq/ha·a or kg/ha·a, Figure 9 E). The grid resolution of the wet deposition map is 1x1km².

Annual wet deposition maps presently are calculated for the years 1987 to 1989 and 1993 to 1995.

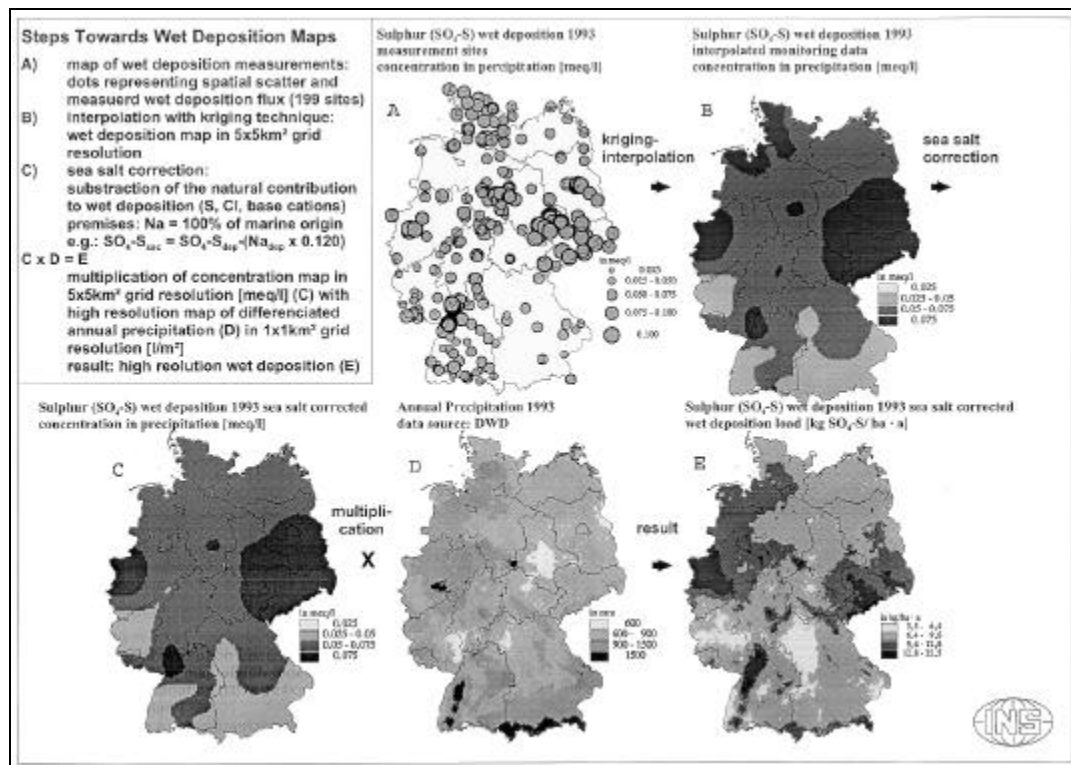


Figure 9: Main procedures of mapping wet deposition loads in Germany

3.1 Sulphur wet deposition (SO₄-S)

The mapping results of sulphur wet deposition (SO₄-S_(ssc)) between 1987 and 1993 show an average decline of about 54% (Figure 10). This clear downward trend reflects more or less the trend of SO₂ air concentration in the same time period (see above). Since SO₄ air concentration, which also contributes to sulphur wet deposition, declined less than SO₂ the reduction of SO₄-S wet deposition shows a less steep declination than SO₂.

The spatial distribution of SO₄-S wet deposition 1987 to 1989 and 1993 to 1995 is shown in Figure 11. In the maps high wet deposition load areas are covering most Germany in 1987. Until 1989 the reduction of the sulphur deposition rates mainly is found in western Germany, whereas in the eastern part to a large extent still shows high values above 750 eq/ha·a (above 12 kg/ha·a respectively). In the years 1993 to 1995 those high deposition areas can only be found at smaller areas around industrialised areas and in higher elevated mountain regions .

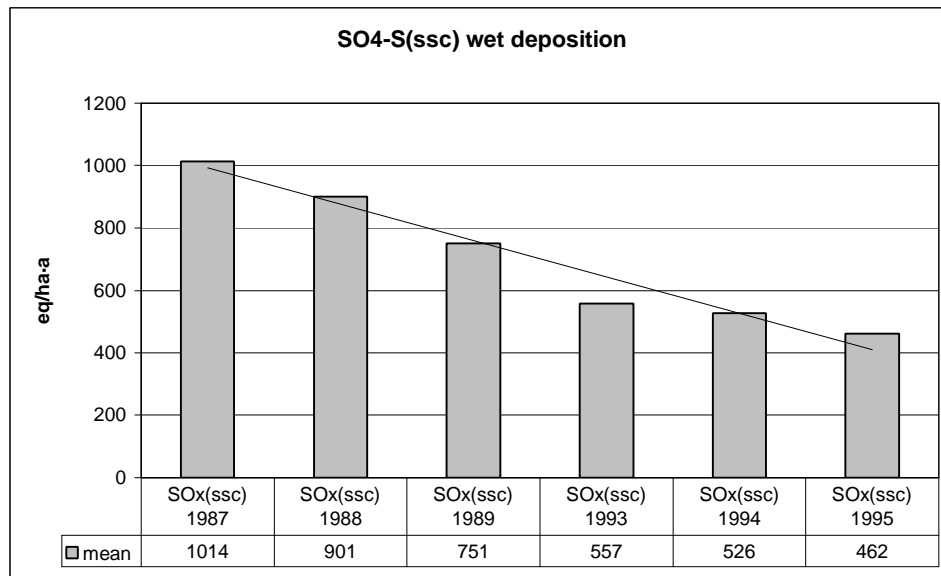


Figure 10: Annual average SO₄-S wet deposition in Germany 1987-1989 and 1993-95

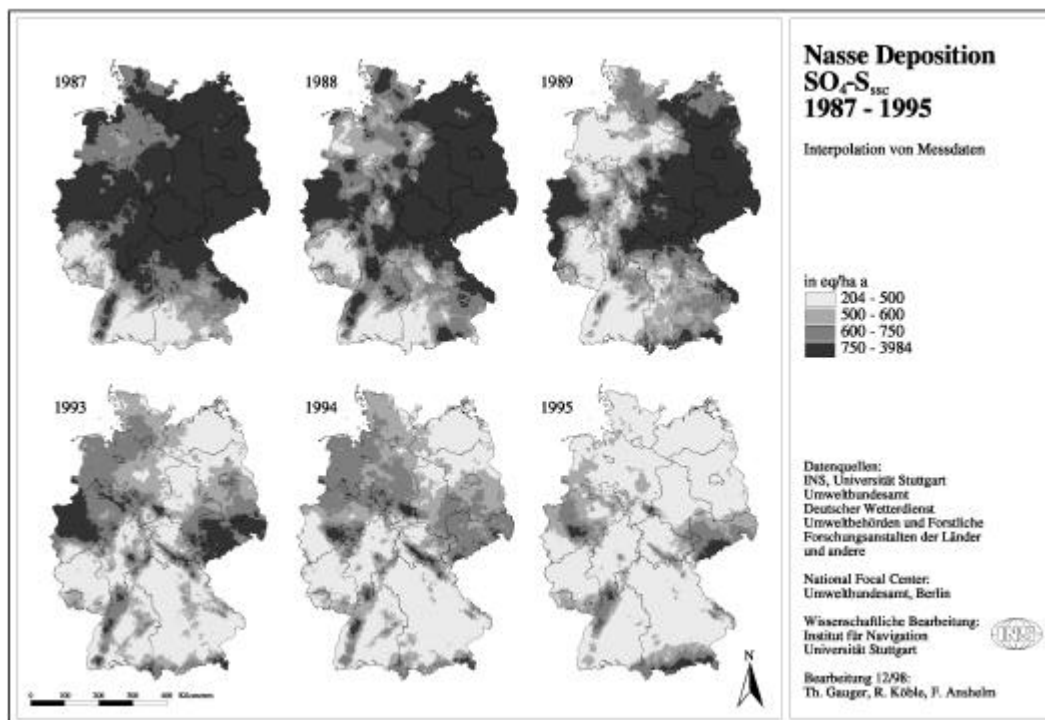


Figure 11: Annual average wet deposition of SO₄-S in the years 1987-89 and 1993-95

3.2 Wet deposition of protons (H)

Wet deposition of protons (H) is used as input for calculating mass loss of unsheltered materials using dose-response functions ('Rain[H⁺]', UBA 1996). The average H wet deposition in Germany declined by about 44% between 1987 and 1995 (Figure 12).

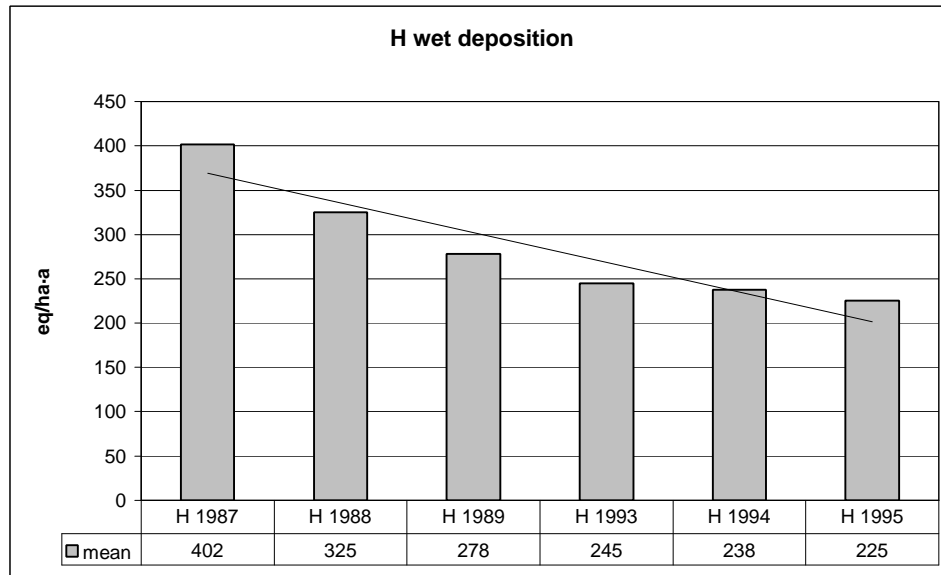


Figure 12: Annual average H wet deposition in Germany 1987-1989 and 1993-95

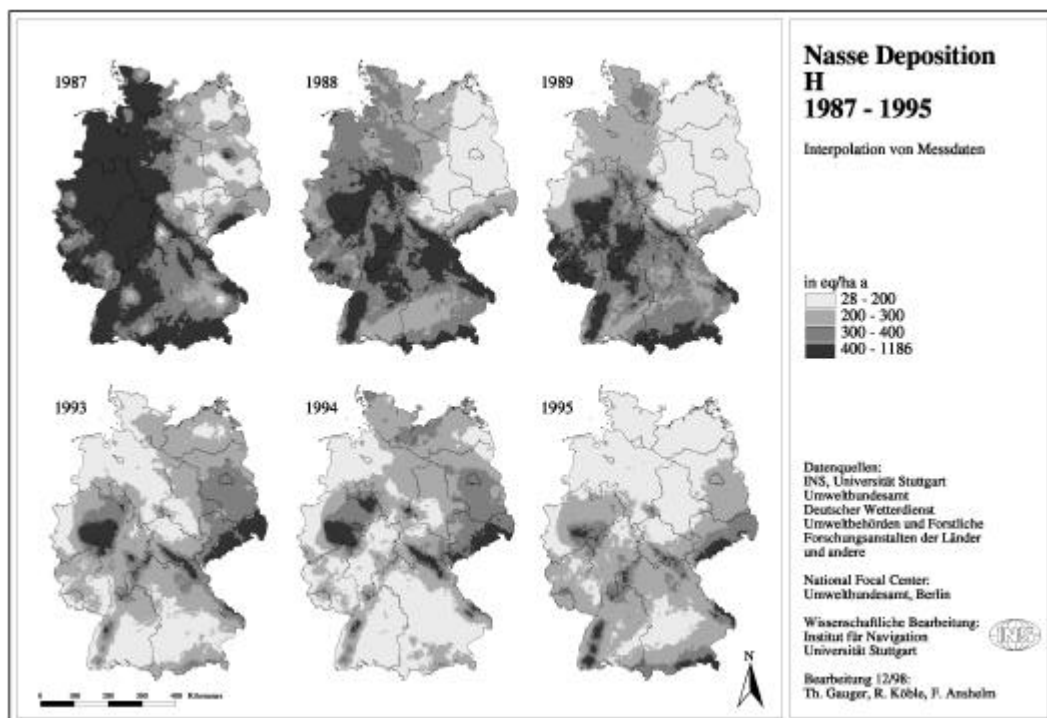


Figure 13: Annual average wet deposition of H in the years 1987-89 and 1993-95

The decline shows no regular spatial trend (Figure 13). Since reactions of solute ions in precipitation are quite complex, wet deposition of protons can be described as a result of the specific mixture and concentration of all deposited ions. The general decline of the wet deposition of hydrogen ions, however, is obvious (Figure 12), though regions with regularly high or low H deposition rates can hardly be identified in the maps (Figure 13).

3.3 Wet deposition of chloride (Cl)

Chloride mainly originates from sea spray, which is transported to inland areas by marine air masses. In Figure 14 the annual average wet deposition of chloride (Cl) in the three year periods 1987-89 and 1993-95 is presented. Cl wet deposition shows a regular gradient from the coastal region of the North Sea (NW) to continental inland Germany (SE). The inland transport distance of marine Cl is connected to the frequency of weather situations with high wind speed, which both leads to higher sea salt emission, and to longer transport distance of marine air masses.

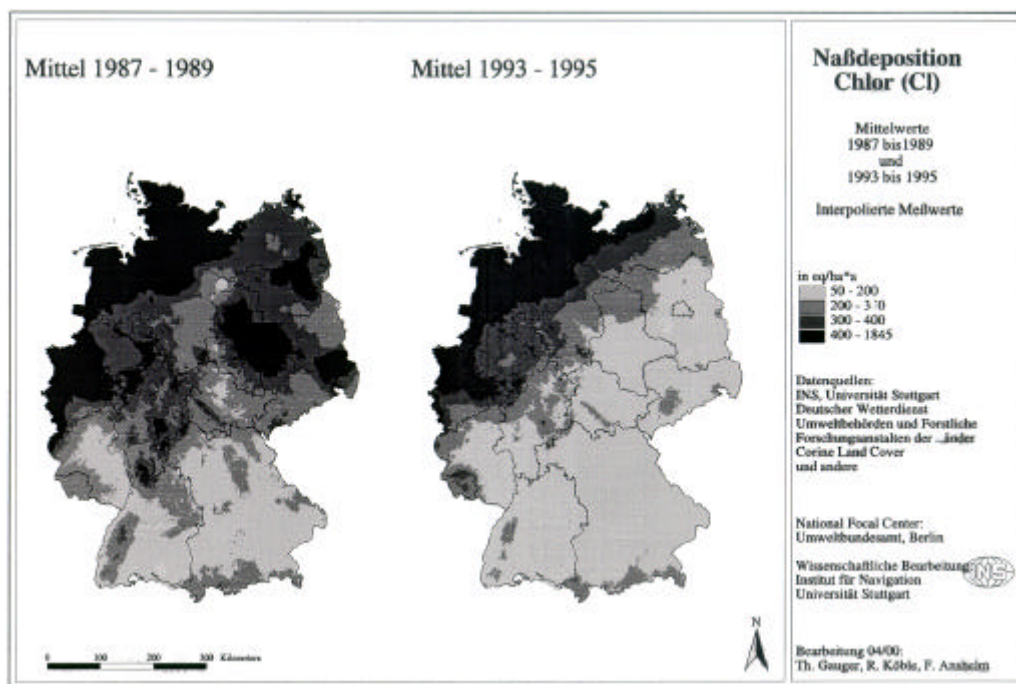


Figure 14: Annual average wet deposition of Cl in the three year periods 1987-89 and 1993-95

The graph in Figure 15 shows the annual variation of the mean Cl deposition rate in the single years 1987 to 1989 and 1993 to 1995. The large interannual variability shows no clear trend, though the average Cl wet deposition rate 1995 is about 13% lower compared to 1987.

The average deposition of non-sea salt chloride ($\text{Cl}_{(\text{ssc})}$) is shown in Figure 16. Here the average wet deposition rate 1995 is about 71% lower compared to 1987. The non-sea salt fraction of Cl is assumed to be completely due to anthropogenic hydrogen chloride (HCl) emissions (UBA 1996, p.31). The ratio between sea salt Cl and total Cl in wet deposition on average rises from 84.6% 1987 to 94.8% 1995. This means that the anthropogenic contribution to total Cl wet deposition declined from about 15% 1987 to 5% in 1995, due to HCl emission reduction (Table 3).

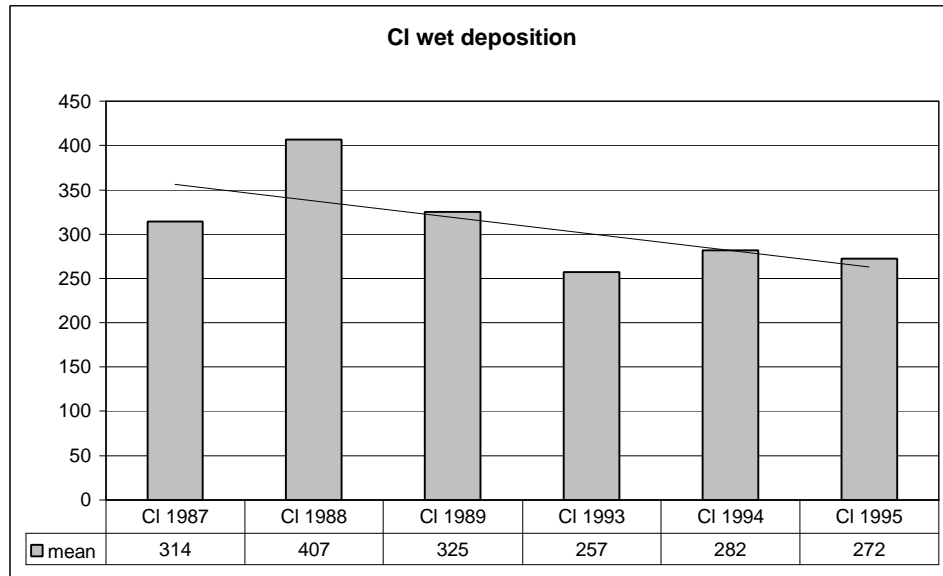


Figure 15: Annual average Cl wet deposition in Germany in the years 1987-89 and 1993-95

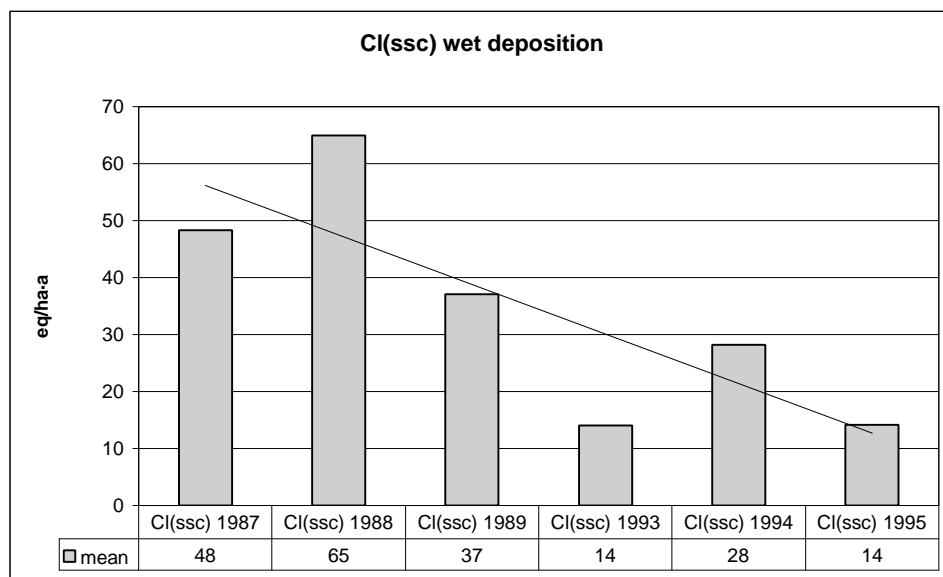


Figure 16: Annual average $\text{Cl}_{(\text{ssc})}$ wet deposition in Germany 1987-1989 and 1993-95

Table 3: Average wet deposition of sea salt chloride

Average wet deposition of sea salt Cl	1987	1988	1989	1993	1994	1995
in eq/ha	265,6	341,8	287,9	242,6	253,3	357,9
in kg/ha	9,42	12,12	10,21	8,60	8,98	9,14
in % of total Cl wet deposition	84,6%	84,0%	88,6%	94,5%	90,0%	94,8%

3.3 Composition of species in wet deposition

The contribution of single ions to the total ion content in wet deposition, due to the magnitude of differences in the temporal and spatial trends, has changed between 1987 and 1995. Figure 17 shows the average contribution of all main components in wet deposition in Germany 1987 (upper graph) and 1995 (lower graph). The contribution of sulphur ($\text{SO}_4\text{-S}$) declines from 32% 1987 to 22% 1995, the contribution of base cations (Ca, Mg, K) declines from 14% 1987 to 10% 1995, whereas the difference in the contribution of H is only about 1%. The contribution of the remaining components is rising, in particular of reduced ($\text{NH}_4\text{-N}$) and oxidised nitrogen ($\text{NO}_3\text{-N}$) from 26% 1987 to 32 % 1995.

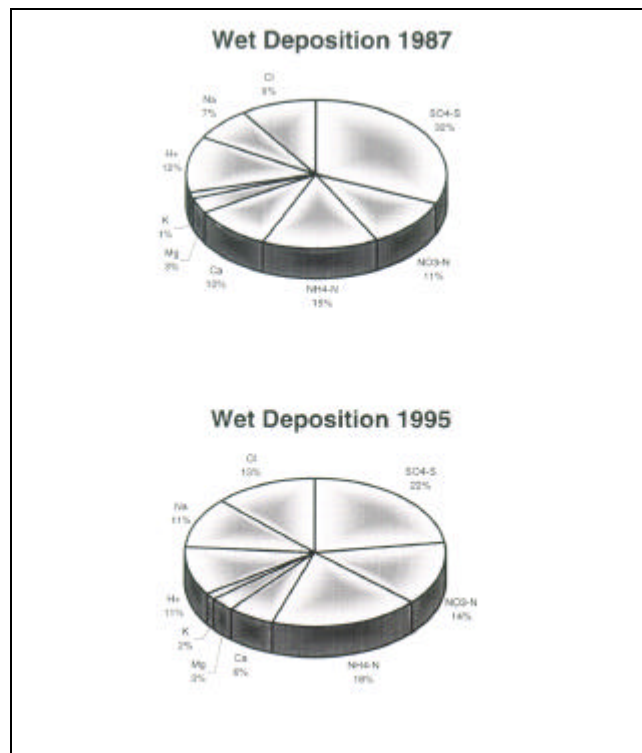


Figure 17: Annual average equivalent distribution in wet deposition 1987 and 1995 in Germany

4. Conclusions

The presented preliminary maps and data of air pollutants are relevant for material deterioration. Changes in the environmental conditions can be observed, in particular the temporal and spatial trends of air concentration and wet deposition of pollutants, due to emission abatement. Data of SO₂ and O₃ air concentration as well as data of chloride concentration in precipitation, Cl wet deposition and wet deposition of H are input parameters of the ICP Materials dose response functions. The experiences made by mapping of actual and acceptable corrosion rates on a national scale in Germany using different dose-response functions showed a strong influence of the spatial differentiation of the input data quality and the spatial trend of the pollutants on the mapping result (see the workshop presentation of Anshelm et al.).

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