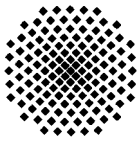

Umwelt Bundes Amt
für Mensch und Umwelt

Final report 299 42 210

Mapping of ecosystem specific long-term trends in deposition loads and concentrations of air pollutants in Germany and their comparison with Critical Loads and Critical Levels

Part 2: Mapping Critical Levels exceedances

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Introduction

Mapping Critical Levels exceedances on the national scale has been one of the major tasks within research project "Mapping of ecosystem specific long-term trends in deposition loads and concentrations of air pollutants in Germany and their comparison with Critical Loads and Critical Levels" (BMU/UBA FE-NO. 299 42 210). Critical Levels exceedance maps as well as maps describing the concentration of air pollutants in Germany have been calculated. Special interest has been put on the detection of long-term trends. Mapping results are based on monitoring data and high resolution land use maps. Regional distinctions in annual air pollutant input to several ecosystems and in Critical Levels exceedances now easily can be located on maps.

One of the most important aims of the project has been support of the German Federal Environmental Agency (UBA) in preparation and review of national datasets to be implemented in European scale Critical Loads and Critical Levels exceedance mapping.

The results of this project have been gained by working in close co-operation with "Gesellschaft für Ökosystemanalyse und Umweltdatenmanagement mbH" (ÖKO-DATA GmbH) located in Strausberg, The Netherlands Organization for Applied Scientific Research (TNO), Appeldoorn and The Netherlands Energy Research Foundation (ECN), Petten, The Netherlands. The research project has been funded by the German Federal Environmental Agency (UBA).

At Institute of Navigation, Stuttgart University (INS), there is a varied experience in mapping Critical Loads & Levels. There are some benefits from advanced knowledge in data handling and mapping procedures. Most useful contacts to experts in manifolds countries now exist for more than ten years. Last but not least, there is a big amount of data on air pollutants stored in INS databases.

INS air concentration database now has been filled up and extended to the years 1996-1999. The database contains data on air concentration of O_3, SO_2, NO, NO_2 and NO_x as well as information on the monitoring sites themselves (chapter 1). For some problems to be solved monitoring sites have to be classified according to their exposition to traffic exhaust gas emissions. The monitoring site classification scheme developed by KÖBLE, GAUGER & ANSHELM (1999) has been modified (chapter 2). There are a lot of rules, regulations and standards to assess air concentration levels of O_3, SO_2, NO, NO_2 and NO_x on the national and/or international scale. Some of them are discussed in chapter 3. Mapping of concentration levels of O_3, SO_2, NO_2 and NO_x has been a major task within this project. Mapping methodology, changes in mapping procedure

as well as mapping results are expatiated in chapter 4. Results of mapping Critical Levels exceedances are described into detail in chapter 5. Additional efforts have been put on the topic ozone and health effects. Exceedances of ozone AOT60 and WHO Air Quality Guideline level for the protection of human health have been calculated and mapped (chapter 6). Finally, there is a contribution to AOT40crops level II mapping. Phenology of wheat has been used as a modifying factor (chapter 7).

1 INS air pollutant concentrations database

A major task within this project has been data acquisition and data preparation for high resolution air pollutant concentrations mapping on the national scale. Data on air pollutant concentrations as well as data on monitoring sites is stored in INS air pollutant concentrations database.

Data on air pollutant concentrations is monitored in stationary monitoring sites. Pursuant to the „Bundes-Immissionsschutzgesetz“ (BImSchG) the German States are under obligation to determine type and extent of air pollution in polluted areas. Data on air pollution in remote background areas is monitored by the monitoring sites of the German Federal Environmental Agency (UBA). Thus, there are 17 monitoring networks within Germany (16 German States + UBA). The German States are responsible for management and maintenance of their monitoring networks. The geographical distribution of the monitoring sites within the monitoring networks is unregular.

The concentration of air pollutants is recorded continuously. After a first check of the quality of the data - carried out by the carriers of the monitoring sites - data on air pollutant concentration is conveyed to German Federal Environmental Agency. Within Federal Environmental Agency data on air pollutant concentrations are harmonised and compiled into a national database. Federal Environmental Agency provides data on air pollutant concentrations to research institutes, for instance Institute of Navigation, Stuttgart University (INS), who are then allowed to use this data for the limited purpose of research. Data are conveyed to INS in ASCII format (0.5- and 1-hour datasets) or in Excel sheets (monthly and/or annual datasets). At INS data on air pollutant concentrations and data on monitoring sites is stored in an Access database.

In previous research projects at Institute of Navigation, Stuttgart university (INS) an air pollutant concentrations database already has been established. This database now has been restructured and extended to the years 1996 to 1999. Datasets of the years before 1996 have been completed. The quality of the data basis used for mapping procedures has improved.

INS air pollutant concentrations database contains data of 1057 monitoring sites (up-to-date August 2001). There is information on the following topics: ID and name of the monitoring sites, coordinates, height above sea-level, commissioning and decommissioning date, address information, type of monitoring site[1] (rural, urban, street, others, no information), some informati

[1] according to Council Directive 92/72/EEC on air pollution by ozone

on about the immediate (0-100m radius) and local environment[2] (100m to some km radius) of the monitoring sites, calibration methods, some extra information on the monitoring sites, results of the INS monitoring site classification scheme described in chapter 2 and, of course, data on concentration of SO_2, NO_2, NO, NO_x and O_3. There is still a lack of information, particularly on the surroundings and the type of the monitoring sites (chapter 2).

Table 1 outlines the kind of air concentration data stored in INS air pollutant concentrations database (up-to-date August 2001). There is 0.5- hourly, hourly, monthly and/or annual data on air pollutant concentrations stored. Data on the concentration of SO_2 and NO_2 is available for the years 1985-1999, data on the concentration of NO for the years 1992-1999 only. Data on the concentration of ozone exists for the years 1988-1999, but the data for the years 1988 and 1989 is incomplete and therefore not suitable for mapping. The data on concentration of NO_2 of the year 1985 also is too patchy for high resolution mapping on a national scale (1 km² grid).

Table 2 indicates data on monitoring sites stored in INS air pollutant concentrations database. Information on ID's (INSCOD, NATCOD) and site name exists for every site. There are still 25 monitoring sites with unknown coordinates, but for these monitoring sites there is no data on air pollutants available anyway. For 919 monitoring sites there is information on its height above sea-level stored in INS air concentration database. Information about the commissioning date of the monitoring sites is available for 857 monitoring sites. From 345 monitoring sites it is known that they are not in commission anymore, because there is information about their decommissioning date. For 418 monitoring sites there is detailed adress information available. For 414 monitoring sites there is information about the type of monitoring site (urban, rural, street), its local and immediate environment, as well as the monitoring and calibration method applied to quantify air pollutant concentration levels. This kind of detailed information is available due to the regulations of the EEC Ozone Directive (92/72/EEC). Finally, for 1021 of 1057 there is information available which is detailed enough to apply the INS monitoring site classification scheme, described in chapter 2.

While integrating new data into INS air pollutant concentrations database existing data on air pollutant concentrations have been checked and validated. With the help of Federal Environmental Agency and the other carriers of monitoring sites (German States) it has been possible to eliminate some discrepancies between old and new datasets, mostly concerning ID´s and/or coordinates.

[2] according to Council Directive 92/72/EEC on air pollution by ozone

Table 1: Kind of air concentration data stored in INS air pollutant concentrations database (1985 – 1999).

gas	averaging time	K[1]	hPa[2]	85	86	87	88	89	90	91	92	93	94	95	96	97	98	99
NO_2	annual mean	293	1013	x	x	x	x	x	x	x	x	x	x	x	x	x	x	x
	monthly mean											x	x	x	x	x	x	x
	hourly mean														x	x	x	x
	0.5-hourly mean											x	x	x				
NO	annual mean										x	x	x	x	x	x	x	x
	monthly mean										x	x	x	x	x	x	x	x
	hourly mean														x	x	x	x
	0.5-hourly mean											x	x	x				
SO_2	annual mean	273		x	x	x	x	x	x	x	x	x	x	x	x	x	x	x
	monthly mean											x	x	x	x	x	x	x
O_3	annual mean	293					x	x	x	x	x	x	x	x	x	x	x	x
	hourly mean								x	x	x	x	x	x	x	x	x	x
	0.5-hourly mean												x	x	x	x	x	x

[1]reference temperature, [2]reference pressure

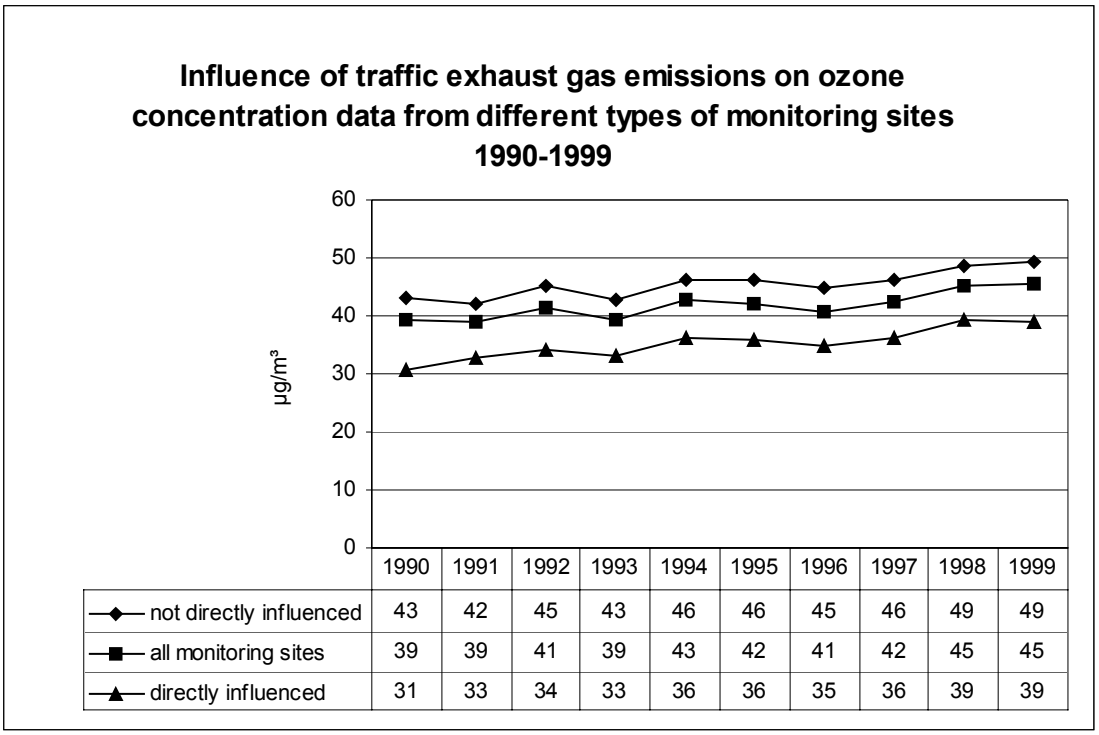
Table 2: Data on monitoring sites stored in INS air pollutant concentrations database.

Metadata information stored in INS air pollutant concentrations database			
	kind of information	**explanation**	**example**
1	INSCOD	numerical code (5-digit, given by INS (first two figures characterize the Bundesland, last three figures characterise the site). INSCOD is directly attached to NATCOD. Numerical codes are easier to handle in ArcInfo.	12053
2	NATCOD	alphanumerical code (5-digit) given by UBA (first two figures characterise the Bundesland, last three figures characterize the site).	BW053
3	SITE	site name	Kuenzelsau
4	RW	northing (7-digit)	3550500
5	HW	easting (7-digit)	5460525
6	LONG_DD	geogr. longitude in decimal degrees	9,69415321
7	LAT_DD	geogr. latitude in decimal degrees	49,28075938
8	ALTITUDE	height above sea-level in meters	214
9	COMM_DATE	commissioning date of the site	01.11.1990
10	DECOMM_DATE	decommissioning date of the site	
11	UNITC	unit of monitoring for the coordinates Longd, -m, -s und Latd, -m, -s	M
12	LONGD	geogr. longitude (degrees)	9
13	LONGM	geogr. longitude (minutes)	41
14	LONGS	geogr. longitude (seconds)	39
15	CARD	position east (E) or west (W) of Greenwich	E
16	LATD	geogr. latitude (degrees)	49
17	LATM	geogr. latitude (minutes)	16
18	LATS	geogr. latitude (seconds)	50,7
19	ADRGE01	adress city	Wertwiesen
20	ADRGE02	adress street	parking place
21	VILLE	adress place	
22	TYPE	type of site R =rural, U = urban, S = street	U
23	IMENV1	code for immediate environment 0 to 100 m radius	I2c
24	IMENV2	code for immediate environment 0 to 100 m radius	I8
25	IMENV3	code for immediate environment 0 to 100 m radius	I99
26	IMENV4	code for immediate environment 0 to 100 m radius	
27	LOCENV1	code for local environment 100 m to some km radius	LE
28	LOCENV2	code for local environment 100 m to some km radius	LH
29	LOCENV3	code for local environment 100 m to some km radius	LJ
30	LOCENV4	code for local environment 100 m to some km radius	LI
31	CODPRIN	monitoring method	UV
32	CALMET	calibration method	E06
33	DIVERS4	additional information ...	-
34	DIVERS5	additional information ...	-
35	CLASSIFICATION	result of INS classification scheme	ok

2 Classifying monitoring sites

The area of representativeness of measured data on air pollutant concentration is heavily influenced by the location of the monitoring sites. There are monitoring sites which are build up to monitor background air pollution levels. These monitoring sites are located in rural areas. Data coming from these monitoring sites have a bigger area of representativeness than monitoring sites located in urban areas or near streets. In particular the sites located near emission sources have a small area of representativeness.

Ozone concentrations are explicitely low near monitoring sites which are directly influenced by traffic exhaust gas emissions (degradation of ozone in the presence of NO, oxidation of NO by ozone, resp.). To map the concentration levels of O_3 and NO_x as well as AOT40, AOT60 and exceedance of WHO Air Quality Guideline Level for ozone the data of street sites should be excluded from the set of data. Figure 1 illustrates the influence of traffic exhaust gas emissions on the concentration of ozone.



	1990	1991	1992	1993	1994	1995	1996	1997	1998	1999
not directly influenced	43	42	45	43	46	46	45	46	49	49
all monitoring sites	39	39	41	39	43	42	41	42	45	45
directly influenced	31	33	34	33	36	36	35	36	39	39

Figure 1: Influence of traffic exhaust gas emissions on ozone concentration data.

A classification scheme has been developed to class monitoring sites according to their exposition to traffic exhaust gas emission sources (KÖBLE, GAUGER & ANSHELM 1999). The classifi

cation scheme allows to select monitoring sites directly affected by traffic exhaust gas emissions, but the classification process has been rather complicated and not all of the criteria used for the classification are still usable when updating to new years. In order to improve the applicability of the classification scheme some modifications have been made. The main goal has been to develop a classification scheme easy to handle, which allows to class the position of a monitoring site to traffic exhaust gas emissions with a high certainty by using only a few criteria (figure 2).

catalog of criteria used for the assessment of the monitoring sites' exposition to traffic exhaust gas emissions*			not affected	directly affected
UBA monitoring site			+	
description of the monitoring site**	TYPE	R		+
		S		+
		U und IMENV1 = I1a, I1b, I2a, I2b, I3a, I3b		+
NO/NO_2 ratio	average 1996-98	>1		+
		<=1	+	

* NO/NO_2 ratio is only used as acriterion, if there is no description of the monitoring site and the site is not a UBA monitoring site.

** for abbreviations look annex 1

Figure 2: Catalog of criteria used for the assessment of the monitoring sites‘ exposition to traffic exhaust gas emissions.

Classification scheme

Monitoring sites should be classified into „urban“, „rural“ or „traffic“ (Annex 1). Traffic sites deliver ozone data with a small radius of representativeness, because of the reaction of O_3 and NO_x. Monitoring sites classified as „urban“ describe the urban background, the rural background air concentration is described by the data of monitoring sites classified as "rural".

The main criterion (criterion 1) for the classification of the monitoring sites is the information delivered by the description of the monitoring sites and their surroundings according to the EEC Ozone Directive (92/72/EEC). Most of the ozone monitoring sites are classified according to this description into urban, rural or traffic. Sometimes there is some information about the local or immediate environment of the monitoring site. Monitoring sites which are located in canyoning streets are classified as directly affected by traffic exhaust gas emissions. The information about the surroundings of the monitoring sites is not always sufficient to get an unmistaken classification. Furthermore, this description is not available for all monitoring sites, but only for

some ozone monitoring sites. Further information sources should be used to get a better idea of the surroundings of the monitoring sites.

The monitoring network maintained by the German Federal Environmental Ageny (UBA) has been established to monitor the air pollution background values within Germany. Therefore, the monitoring sites of this monitoring network lie in rural surroundings, far away of the sources of traffic exhaust gas emissions. All monitoring sites of the Federal Environmental Agencies monitoring network are classified as rural (criterion 2).

If there is no description of the sites‘ surroundings available and the site is not a monitoring site of the Federal Environmental Agencies‘ monitoring network the NO/NO_2 ratio is taken as a criterion (criterion 3). Near streets the NO/NO_2 ratio is high, because of the traffic exhaust gas emissions. Cars emit more than 95 % of their total nitrogen oxides exhaust gases as NO (BAUMBACH 1994: 107-110; LEISEN 1992: 266). In ambient air NO is oxidised to NO_2 very quickly (within seconds). Therefore there is a gradient from high NO/NO_2 ratios nearby streets to low NO/NO_2 ratios far from traffic exhaust gas emissions.

An internal study of the German Federal Environmental Agency suggests a NO/NO_2 ratio of > 1 as an indicator for proximity to traffic exhaust gas emission sources. The study is based on data of the German NO and NO_2 monitoring sites of the years 1996-1998 (oral information Ms. Grittner/Mr. Breuninger from Federal Environmental Agency, Berlin). This value (NO/NO_2 ratio >1 = directly affected by traffic exhaust gas emissions) has been verified on the basis of the data of the INS air pollutants concentration database.

To check the results of the internal UBA study the NO and NO_2 data of the years 1996-1998 have been used to calculate NO/NO_2 ratios. The results have been compared with the description of the monitoring sites. There is no UBA monitoring site and only one rural site with a NO/NO_2 ratio > 1. The level of the NO/NO_2 ratio of traffic related monitoring sites is obviously higher than the NO/NO_2 ratio of the other monitoring sites (figure 3).

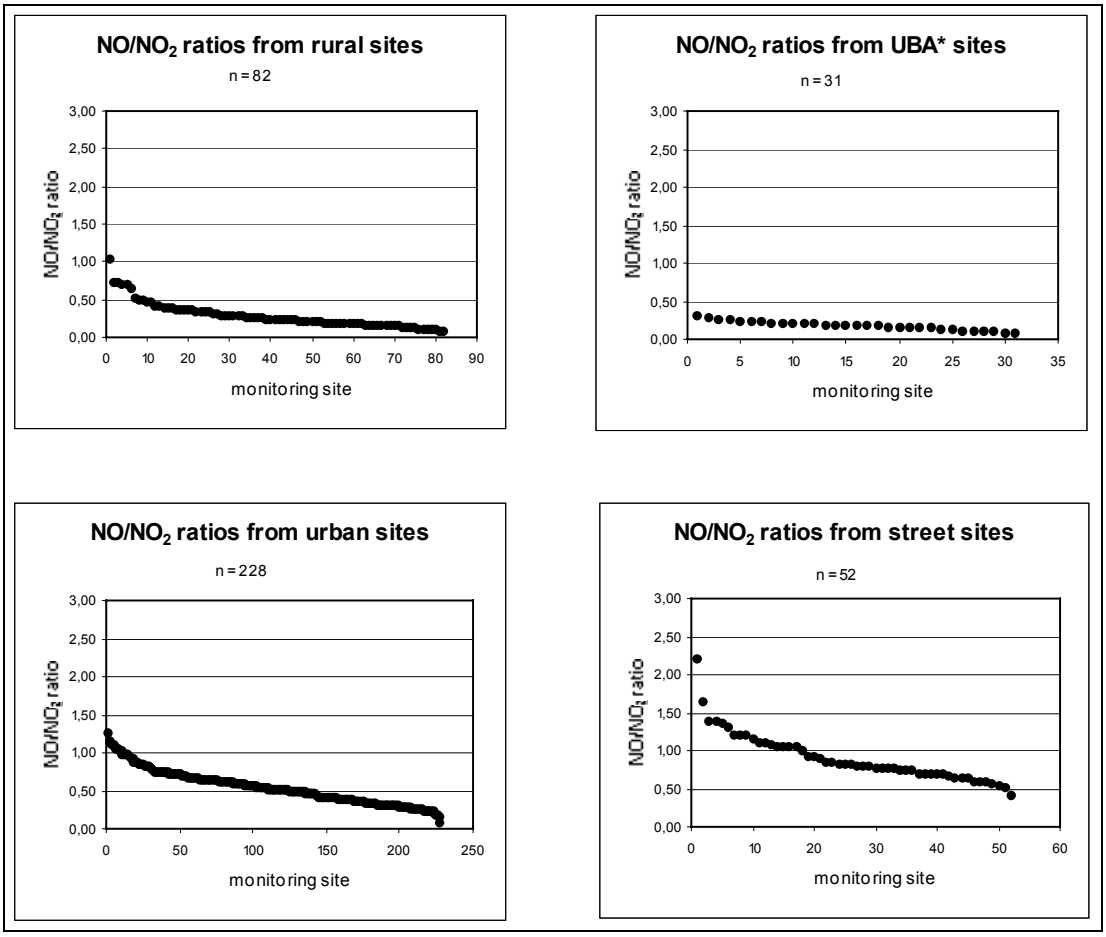


Figure 3: NO/NO_2 ratios from rural, UBA*, urban and street sites (*UBA = Federal Environmental Agency monitoring network = rural background).

Finally, it has been decided to use the NO/NO_2 ratio of 1 as a criterion to distinguish between the monitoring sites directly affected by traffic exhaust gas emissions (NO/NO_2 ratio >1) and the monitoring sites not directly affected by traffic exhaust gas emissions (NO/NO_2 ratio ≤1).

Table 3 indicates minima, maxima and mean values of the NO/NO_2 ratio of all monitoring sites, the sites of the UBA monitoring network, as well as of the monitoring sites classified as „rural“, „urban“ or „traffic“. Highest values can be found at traffic related sites, lowest values at UBA monitoring sites. The category „all monitoring sites“ includes the monitoring sites without any description of the site.

Table 3: Minimum, mean and maximum NO/NO_2 ratios from different sets of monitoring sites.

NO/NO_2 ratio	minimum	mean	maximum
all sites	0.07	0.63	2.67
UBA sites	0.08	0.18	0.31
rural sites	0.08	0.28	1.05
urban sites	0.07	0.54	1.24
street sites	0.4	0.9	2.21

The classification of the monitoring sites is performed between the wish to exclude all data of monitoring sites directly influenced by traffic exhaust gas emissions and the necessity to have an adequate density of the monitoring network. When using the criterion of a NO/NO_2 ratio of > 1 only the heavy affected monitoring sites get excluded. This guaranties a high density of measured air concentration data for the following interpolation process. The lower the density of the monitoring network and the more unregular the distribution of the monitoring sites, the lower is the trustiness of the interpolated values. The argument to have a high density of the monitoring network has been weighted higher than the wish to exclude with highest certainty the data of monitoring sites directly affected by traffic exhaust gas emissions. Fundamentally, the line between directly and not directly affected by traffic exhaust gas emissions can not be drawn at a specific limit. The peculiarities of the site specific factors (covering with buildings, vegetation, traffic density, habit of the site, air circulation, climatic parameters) are too manifold.

Factors to be considered in the revised classification scheme (Annex 1 and figure 2)

- all monitoring sites of the type „R“ = rural (extended description of the monitoring sites) pass for not directly affected by traffic exhaust gas emissions. These monitoring sites are used for the interpolation and mapping process

- all monitoring sites of the type „S“ = street (extended description of the monitoring sites) pass for directly affected by traffic exhaust gas emissions. These monitoring sites are not used for the interpolation and mapping process

- out of the pool of monitoring sites of the type „U“ = urban (extended description of the monitoring sites) these are not used which lie at

    - big heavy or medium frequented streets (ImEnv = I1a, I1b)[3]

    - small heavy or medium fequented streets (ImEnv = I2a, I2b)

- heavy or medium frequented canyoning streets (ImEnv = I3a, I3b)

- monitoring sites of the UBA monitoring network are used for interpolation and mapping process

- all monitoring sites with a NO/NO_2 ratio > 1 are not used for interpolation and mapping process

The modified classification scheme should need as less data as possible, should be easy to use and should be as clear as possible in its classification. The number of possible categories has been reduced to three (urban, rural, street). Altogether it has been possible to class more monitoring sites than when using the old classification scheme.

Up to date air pollutant concentration data of 1054 monitoring sites are available. Hereof 203 monitoring sites are classified as directly affected by traffic exhaust gas emissions when using the new classification scheme. All monitoring sites, which are classified as directly affected by traffic exhaust gas emissions are excluded from the interpolation and mapping of the concentration of O_3 and NO_X, as well as of mapping AOT40, AOT60, exceedance of WHO Guideline level for the protection of human health.

[3] for list of codes see Annex 1

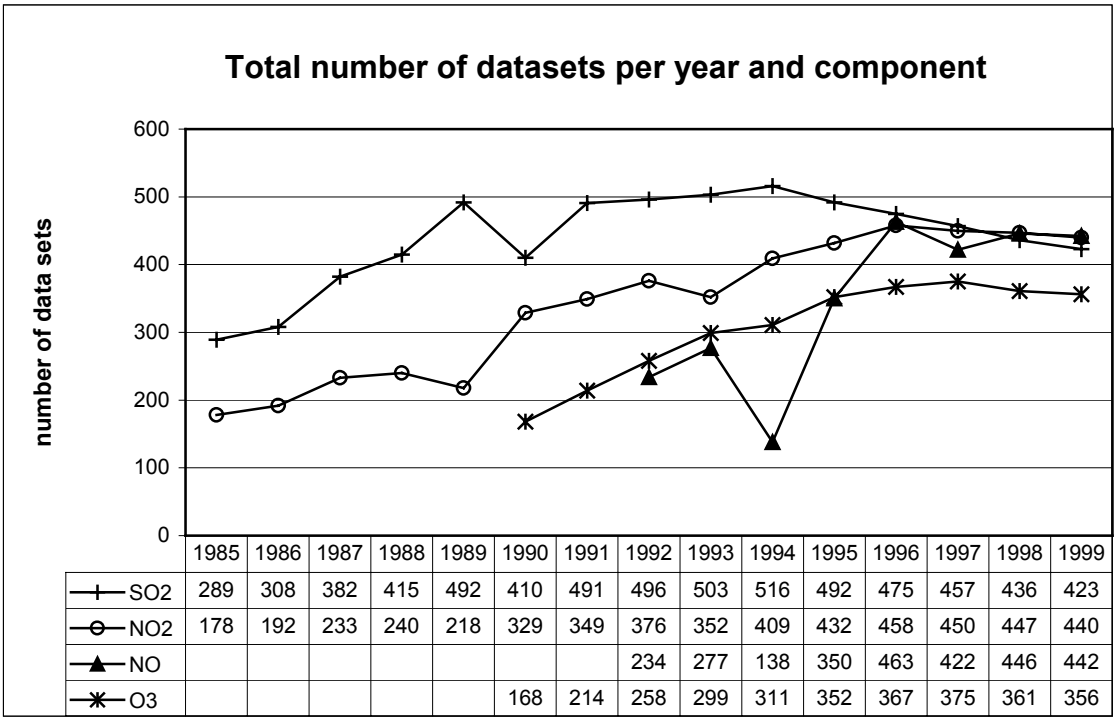
3 Mapping air pollutant concentrations

Mapping air pollutant concentrations on the national scale has been one of the major tasks of this project. The final goal of the mapping process is to calculate maps, which supply information about the concentration of SO_2, NO_x and O_3 for all points offside the monitoring sites of the German air pollutant concentrations monitoring networks. Kriging techniques are applied to calculate these maps.

The quality of the interpolated data is determined by the quality of the measured data themselves and in particular by the quality of the underlying point data set. A high density of monitoring sites and an evenly distributed sample of monitoring data favour a good interpolation result (i. e. low Kriging variances). In areas where the density of the monitoring network is low the spatial reliability of the interpolated data is low. The quality of the underlying monitoring networks should be kept in mind when interpretating the following maps on air pollutant concentrations and Critical Levels exceedances. The number of monitoring sites and the spatial distribution of the monitoring sites varies from year to year and pollutant to pollutant. Therefore in the following sub-chapters the interpolation results are discussed together with the underlying set of monitoring sites.

Figure 4 indicates the number of datasets (monitoring sites with data on air pollutant concentrations) per year and component stored in INS air pollutant concentrations database. INS air pollutant concentrations database contains data of the years 1985 to 1999. The total number of datasets for SO_2 has increased from 289 to 423 between 1985 and 1999. In 1990 there has been a short-time decrease in the number of SO_2 datasets. This irruption is due to the political changes during the German Unification. Maximum number of SO_2 datasets is available for the year 1994 (516). Till the end of the 1990ies the number of SO_2 monitoring sites slowly decreased, because due to the success of emission abatement measures monitoring of the concentration of SO_2 became less important. The total number of datasets for NO_2 has increased from 178 to 440 between 1985 and 1999. In the German Democratic Republic the concentration of nitrogen oxides has been monitored in exceptional cases only. Since 1990 the concentration of nitrogen oxides is monitored both in the western and the eastern parts of Germany. The maximum number of NO_2 datasets has been reached in 1996 (458). The series of monitoring data on the concentration of NO starts in 1992. The total number of datasets for NO increased from 234 in 1992 to 442 in 1999. In 1994 there is a sharp fall in number of NO datasets. This is due to a data crash in Federal Environmental Agency‘s database. The maximum number of NO datasets has been reached

in 1996 (463). The series of data on the concentration of ozone starts in 1990. The total number of O_3 datasets increased from 168 in 1990 to 356 in 1999. The maximum number of O_3 datasets (375) has been available in 1997.



	1985	1986	1987	1988	1989	1990	1991	1992	1993	1994	1995	1996	1997	1998	1999
SO2	289	308	382	415	492	410	491	496	503	516	492	475	457	436	423
NO2	178	192	233	240	218	329	349	376	352	409	432	458	450	447	440
NO								234	277	138	350	463	422	446	442
O3						168	214	258	299	311	352	367	375	361	356

Figure 4: Total number of datasets per year and per component.

For ozone and nitrogen oxides the total number of available datasets is not identical with the number of datasets suitable for mapping (table 4). The data of monitoring sites directly influenced by traffic exhaust gas emissions are not suitable for mapping on the national scale and therefore have to be removed. The procedure how to identify monitoring sites directly influenced by traffic exhaust gas emissions is described in chapter 2. Table 4 indicates the number of datasets suitable for mapping.

Table 4: Number of datasets suitable for mapping per year and component.

Number of datasets suitable for mapping per year and component															
	1985	**1986**	**1987**	**1988**	**1989**	**1990**	**1991**	**1992**	**1993**	**1994**	**1995**	**1996**	**1997**	**1998**	**1999**
SO_2	289	308	382	415	492	410	491	496	503	516	492	475	457	436	423
NO_2	95*	104	131	136	132	195	201	217	205	242	250	275	248	254	239
NO	-	-	-	-	-	-	-	108	142	78*	189	274	239	254	238
O_3	-	-	-	-	-	110	133	156	173	187	212	223	229	227	219
AOT40c	-	-	-	-	-	107	133	156	177	189	210	224	228	230	221
AOT40f	-	-	-	-	-	109	135	158	176	189	212	225	230	227	222
WHO120	-	-	-	-	-	119	153	168	189	203	216	231	231	243	230
AOT60	-	-	-	-	-	110	133	156	173	187	212	223	229	227	219

*number of datasets not sufficient for mapping

3.1 Mapping concentration of SO_2

Sulfur dioxide is predominantly a product of combustion processes. Sulfur, which is included in the fuel gets oxidised during combustion and is emitted in the atmosphere. Process-related emissions of SO_2 occur in particular in the field of conditioning crude oil or natural gas, in metal production and in chemical industry.

Sulfur dioxide is known as a corrosive gas for a long time. It has negative effects on human health, plants and materials. SO_2 affects plants and materials by forming sulfuric acid („acid rain“) and human health by irritating the mucous membranes of the respiratory organs causing respiratory problems.

There are numerous regulations both on the national and international level dealing with emissions and concentrations of SO_2. Figure 5 gives a review of some of the more recent regulations dealing with emissions and concentrations of SO_2. Beyond the regulations mentioned in figure 5 there has been another important regulation exclusively dealing with the emission rates of SO_2 - the German Großfeuerungsanlagenverordnung of 22 June 1983 (13. BImSchV). The 13.BImSchV has had a wide influence on the emission and concentration of SO_2 in Germany. The regulation aggravated the emission threshold values for power plants. The carriers of the power plants had to put the regulations of the 13.BImSchV into action within a time limit of five years. These measures were restricted to the old West German States until the political changes at the beginning of the 1990ies. After the German Unification the newly-formed German States had to implement the regulations of the 13.BImSchV, too.

regulation	component	value	unit	averaging time	aspect	character	comments
WHO (1999)	SO_2	500	µg/m³	10-min average	human health	guideline value	
		125		24-h average			
		50		annual average			
		100		24-h average	vegetation	guideline value	
		30		annual average			
1. Daughter Directive (1999/30/EC) to Air Quality Framework Directive (96/62/EEC)	SO_2	350	µg/m³	1-h average	human health	threshold value	no more than 24 exceedances per year permitted (target year: 2005)
		125		24-h average			no exceedances permitted (target year: 2005)
		20		annual average	ecosystems		no exceedances permitted
UN ECE "Gothenburg protocol"	SO_2	10	µg/m³	annual average	lichens	critical level	
		20			forests and natural vegetation		
		30			crops		
TA Luft (1.BImSchV)	SO_2	140	µg/m³	annual average	human health	threshold value	IW1 = long-term effects
		400		98 percentile of the cumulative frequency of the 0.5-h averages of one year			IW2 = short-term effects
22.BImSchV (89/427/EEC, 80/779/EEC)	SO_2	80	µg/m³	median of 24-h averages of one year[1]	human health and environment	threshold value	
		120		median of 24-h averages of one year[2]			
		250		98 percentile of the cumulative frequency of 24-h averages of one year[3]			
		350		98 percentile of the cumulative frequency of 24-h averages of one year[4]			

[1] if airborne particles > 150 µg/m³ [2] if airborne particles <= 150 µg/m³ [3] if airborne particles > 350 µg/m³ [4] if airborne particles <= 350 µg/m³

Figure 5: Air quality standards for the concentration of SO_2.

In consequence of the regulations mentioned above various measures have been taken to abate SO_2 emissions. As figure 6 and 7 indicate, the measures met with some success. Between 1980 and 1985 total emission of SO_2 in Germany accounted for about 7300 to 7700 kilotons per year (figure 6). The maximum emission rate has been reached in 1985 (7732 kilotons). Since 1986 the annual emission rate of SO_2 decreases. Between 1985 and 1998 the total emission of SO_2 in Germany decreased by 83.2 %. In 1998 the total emission amounted to 1292 kilotons. For 1999 emission data are not yet available.

Figure 7 indicates the differences in emission rates between the old West German States and the newly-formed German States. Since 1986 there is a stable downward trend in emission rates both in the old West German States and in the newly-formed German States. In the single years approximately 72 % of the emissions of SO_2 account for the newly-formed German States, 28 % can be assigned to the old West German States. At the beginning of the 1990ies the political and economic changes lead to a striking reduction of the emission rates in the newly-formed German States. Within one year (1990/1991) emission rates decreased by almost 1000 kilotons. In 1998 the emission rates in the newly-formed German States are still much higher then those in the old West German States.

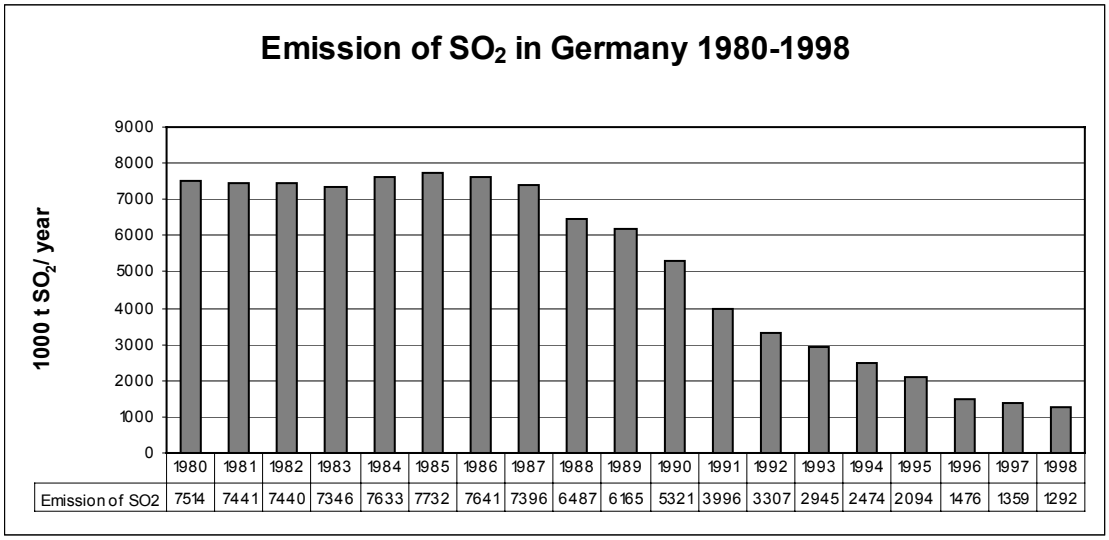


Figure 6: Emission of SO_2 in Germany 1980–1998 (EMEP/MSCW 2000).

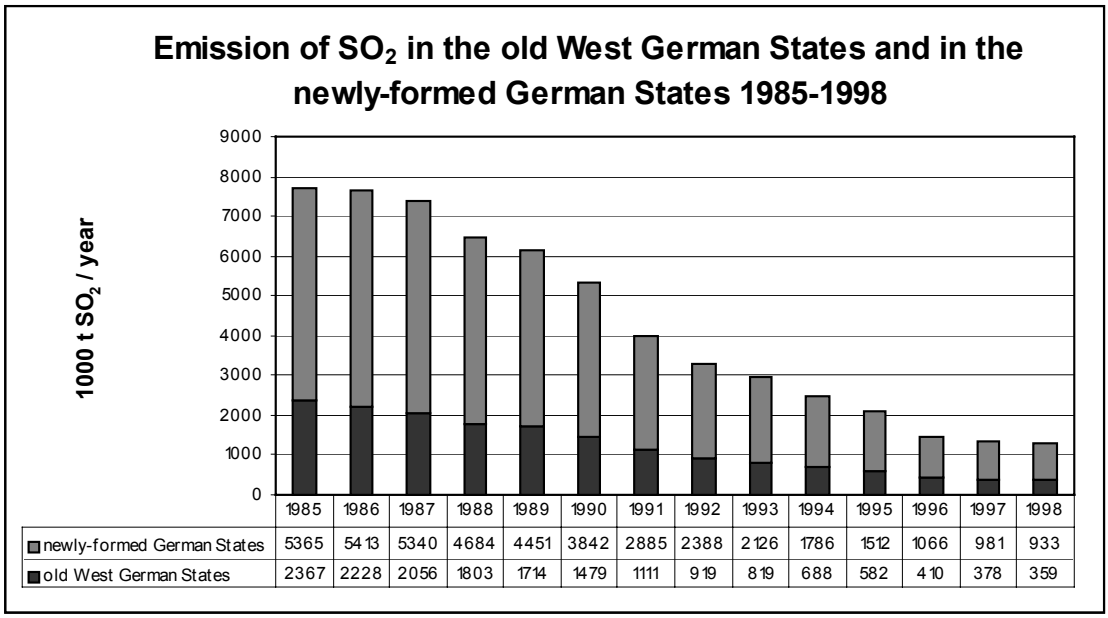


Figure 7: Emission of SO_2 in the old West German States and the newly-formed German States 1985–1998 (EMEP/MSCW 2000).

Mapping of the concentration of SO_2 has been performed for the years 1985-1999. Map 1 illustrates the development of the SO_2 monitoring network. In 1985 there had been rather big gaps in the monitoring network. Only in highly industrialised zones (Ruhr Basin, along the Rhine, Berlin, Hamburg) and in Saxony there has been a high density of monitoring sites. From 1985 til 1989 there has been a steady rise of the number of SO_2 monitoring sites (table 4). In 1985 data of 289 monitoring sites have been available. Till 1989 more and more monitoring sites have been built up. In 1989 there are 492 datasets available. The density of the monitoring network has increased considerably. In 1990 (410 datasets) there has been a short-time irruption, mainly because of the political changes in the German Democratic Republic, but also because of a lack of information about the monitoring sites in Rhineland-Palatinate and Saarland. In 1991 the number of monitoring sites again has reached the number of monitoring sites already existent in 1989. In 1994 the number of monitoring sites measuring air concentration of SO_2 reached its peak (516). Since 1995 there is a decrease in the number of monitoring sites measurering SO_2, but in 1999 there still has been SO_2 concentration data of 423 monitoring sites available. This decrease may reflect the decreasing importance of the parameter SO_2 within the mixture of air pollutants. But the number of monitoring sites and the distribution of the monitoring sites within Germany is still quite good.

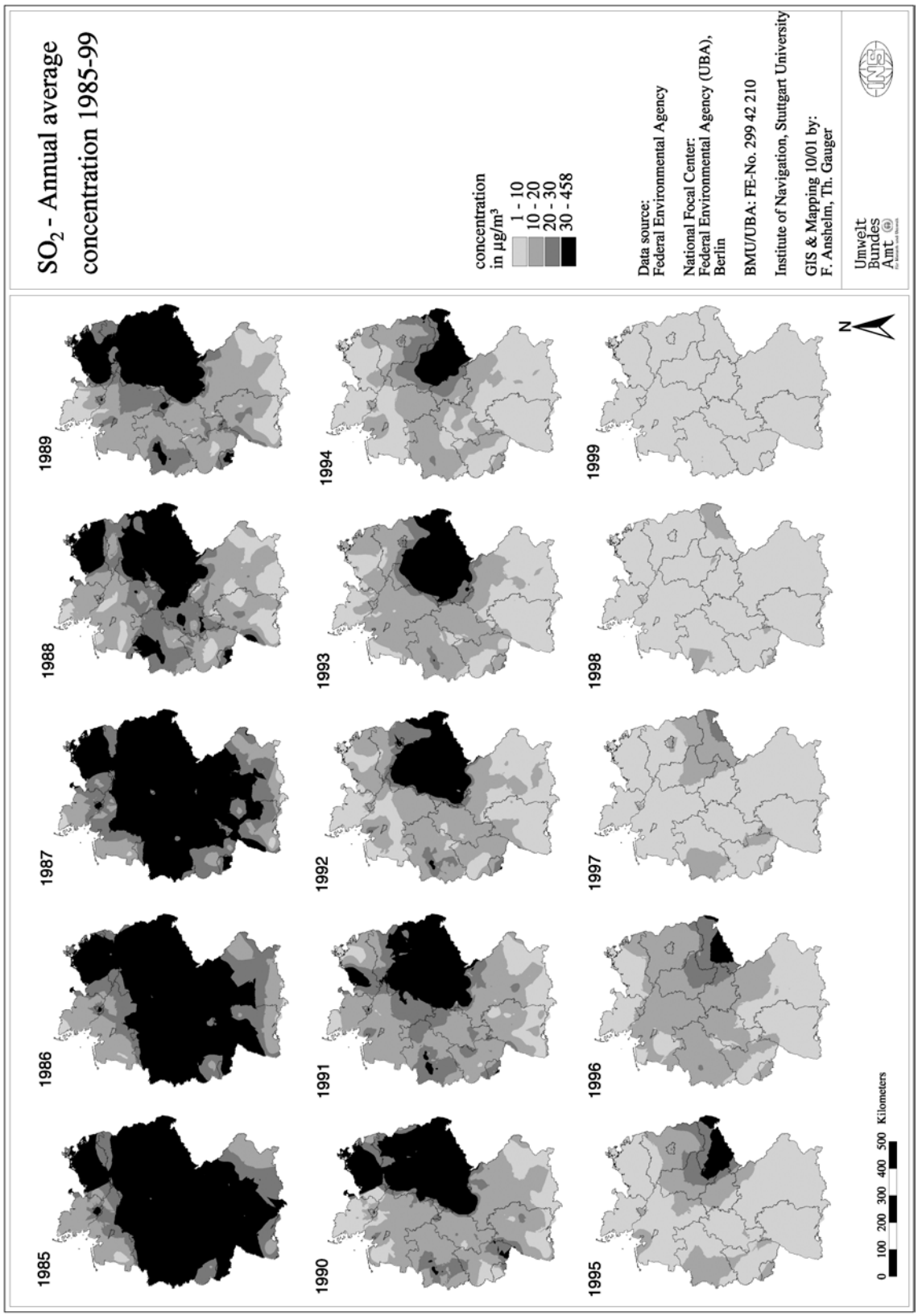
Map 2 indicates the mapping results for the years 1985-1999. There is a downward trend of the annual average concentration of SO_2 in Germany (figure 8). In 1985 the annual average air concentration of SO_2 within Germany has been 52 µg/m³. In 1999 the annual average calculated for the whole area of Germany is 5 µg/m³. This corresponds to a decrease of about 90 %. In every single year there is, of course, a wide range of annual average SO_2 concentrations throughout Germany. In 1987 the range of annual average SO_2 concentrations has been widest. There have been values between 5 µg/m³ (Garmisch-Partenkirchen) and 458 µg/m³ (southern Saxony-Anhalt). This range decreased rapidly after German Unification (1989/1990). In 1999 the annual average SO_2 concentration varied between 2 µg/m³ (Mecklenburg-Western Pomerania, Schleswig-Holstein, SE-Bavaria) and 19 µg/m³ (Ruhr Basin, Saarbrücken).

The reduction of SO_2 concentration neither took place continuously nor in every place simultaneously. There are regional differences visible and in a single year (1996) there even is a deviation from the long-term trend (map 2).

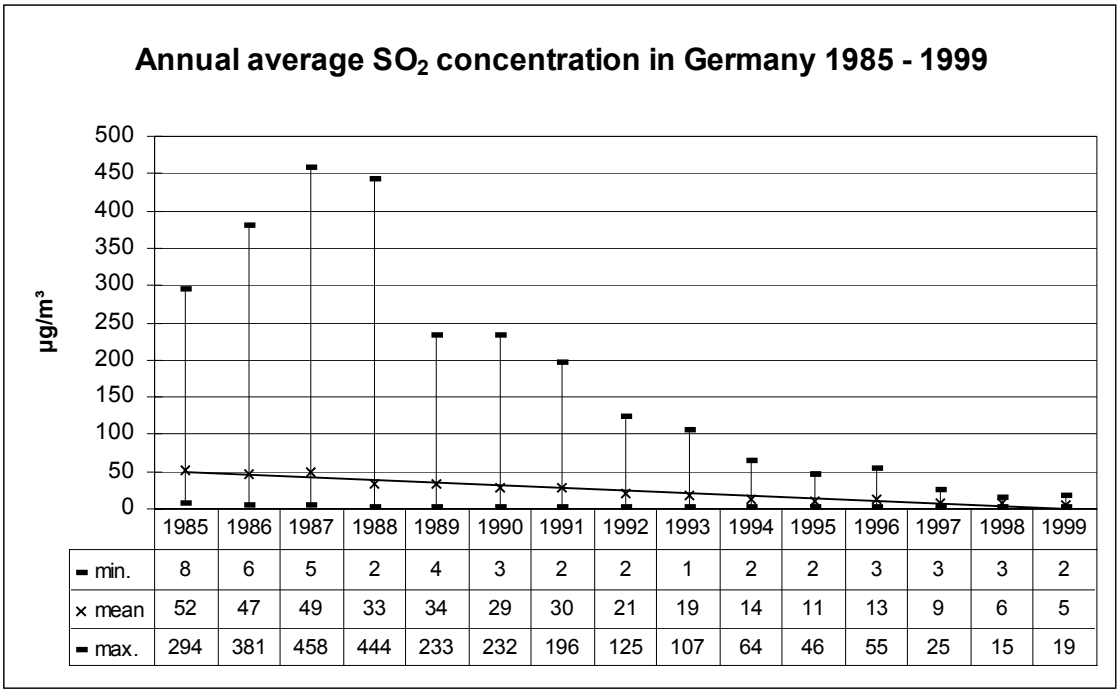
The annual average concentration of SO_2 is influenced by the emission situation, long-range transport processes and the weather conditions especially in wintertime. This interaction between weather conditions and emission reduction measures explains the striking reduction of the concentration of SO_2 within the old West German States between 1987 and 1988 (map 2 and figure 9). Between 1985 and 1987 the effects of the emission reduction measures had been masked by weather conditions. The years 1985, 1986 and 1987 had been explicitely too cold compared to the long-term annual average temperature in Germany (figure 10). A longer and more intensive firing period as well as a relatively high frequency of occurrence of inversions (reduced exchange of air masses) lead to high concentrations of SO_2, despite the emission reduction measurements. At the end of the 1980ies and the first half of the 1990ies annual average temperatures in Germany had been higher than the long-term average. Especially in wintertime the average temperature had been relatively gentle in these years (figure 10). This lead - in combination with the effects of the emission reduction measures – to a striking reduction of the emission rates and concentrations of SO_2 (FRICKE & UHSE 1994) (map 2).

Within the newly-formed German States the decline of the concentration of SO_2 set in some years later than in the old West German States, but at the end of the 1990ies the level of concentration of SO_2 has equalized (map 2 and figure 9). 1999 has been the first year, in which the annual average concentration of SO_2 of the newly-formed German States lies beyond the annual average SO_2 concentration of the old West German States.

Map 1: SO_2 – Distribution of monitoring sites 1985-99.

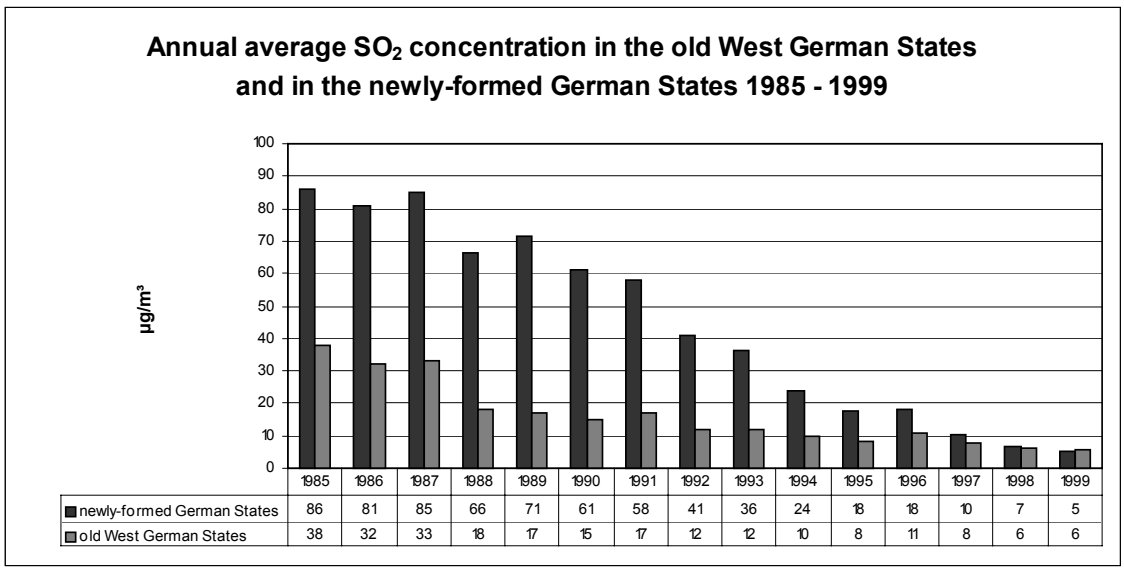


Map 2: SO_2 – annual average concentration 1985-99.



	1985	1986	1987	1988	1989	1990	1991	1992	1993	1994	1995	1996	1997	1998	1999
min.	8	6	5	2	4	3	2	2	1	2	2	3	3	3	2
× mean	52	47	49	33	34	29	30	21	19	14	11	13	9	6	5
max.	294	381	458	444	233	232	196	125	107	64	46	55	25	15	19

Figure 8: Annual average SO_2 concentration in Germany 1985-1999 (EMEP/MSCW 2000).

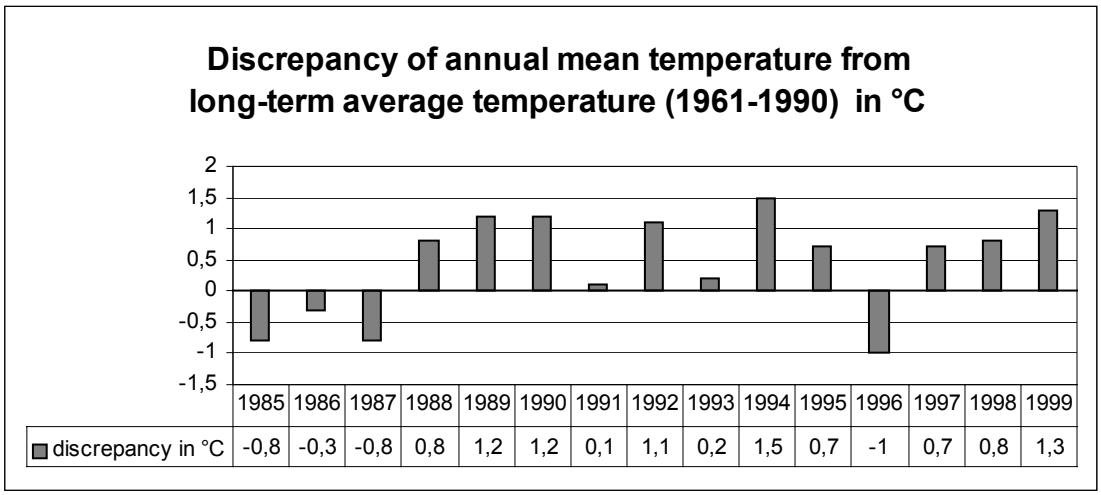


	1985	1986	1987	1988	1989	1990	1991	1992	1993	1994	1995	1996	1997	1998	1999
newly-formed German States	86	81	85	66	71	61	58	41	36	24	18	18	10	7	5
old West German States	38	32	33	18	17	15	17	12	12	10	8	11	8	6	6

Figure 9: Annual average SO_2 concentration in the old West German States and in the newly-formed German States 1985-1999.

A slight deviation from the long-term downward trend in concentration of SO_2 occurred in 1996. There has been a slight increase in comparison to the preceding year, because of the weather conditions in winter time. January (-2.5 °C), February (-2.5 °C), March (-2.7 °C) and December 1996 (-3.2 °C) had been explicitely too cold in comparison with the long-term avera

ge temperature (1961-1990). On the whole the 1996 annual average temperature had been 1.0 °C too cold (figure 10) (ULLRICH, RAPP & FUCHS 2000). A longer and more intensive firing period as well as a high frequency of occurrence of inversions lead to relatively high concentrations of SO_2. In the following years the long-term trend of decreasing concentrations of SO_2 continued. In 1999 the annual average concentration of SO_2 in Germany accounted for 15-20 µg/m³.



	1985	1986	1987	1988	1989	1990	1991	1992	1993	1994	1995	1996	1997	1998	1999
discrepancy in °C	-0,8	-0,3	-0,8	0,8	1,2	1,2	0,1	1,1	0,2	1,5	0,7	-1	0,7	0,8	1,3

Figure 10: Discrepancy of annual mean temperature from long-term average temperature (1961-1990) in °C (Ullrich, Rapp & Fuchs 2000).

3.2 Mapping concentration of nitrogen oxides (NO_2, NO)

Nitrogen oxides (NO_2, NO) are chemical byproducts of high temperature combustion processes, mainly in automobiles and in combustion plants (oxidation of nitrogen included in the fuel and in the combustion air). Chemical industry (e. g. production of fertilizers, production of nitric acid) is another source of nitrogen oxides. Nitrogen oxides are emitted primarily as NO. In the atmosphere NO is rapidly oxidised to NO_2 by oxygen and in particular by ozone. Within a couple of minutes there is a equilibrium between NO, NO_2 and O_3. This equilibrium is on the side of NO_2 as long as the concentration of NO_x is low, the concentration of O_3 is high or radiation is high. This means, under these conditions the concentration of NO_x is dominated by NO_2 and only a small share of NO_x is made up by NO (UBA 1999:33). Close to emission sources the share of NO may be higher than the share of NO_2. Aside from emission sources the concentrations of NO are not noteworthy (UBA 1999:127). Mapping activities therefore are limited to the concentration of NO_2 and the concentration of NO_x. NO_x is a umbrella term for the sum of the concentration of NO_2 and the concentration of NO calculated as specified below.

$$NO_x\ [\mu g/m^3] = (NO_2[\mu g/m^3] / 2.05 + NO[\mu g/m^3] / 1.34) \cdot 2.05.$$

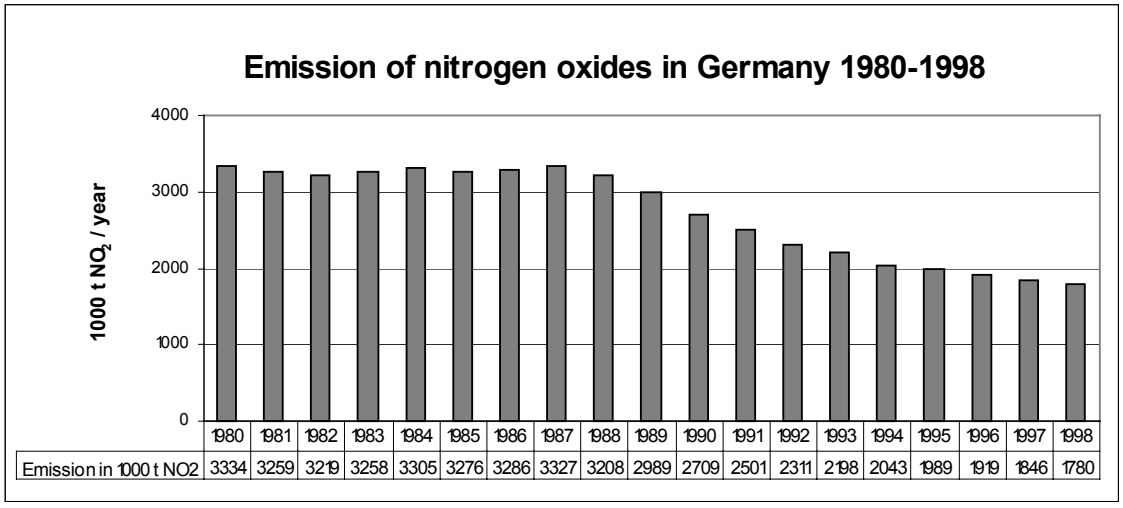
Nitrogen oxides irritate the mucous membranes of the respiratory organs and favour respiratory problems. Beyond it, nitrogen oxides are acidifiers (nitric acid) and therefore can cause damage to plants, buildings and monuments („acid rain"). Gaseous nitrogen oxides can also have a fertilizer effect to plants. Therefore nitrogen oxides can also contribute to over-fertilization of our ecosystems. After all, nitrogen oxides influence the concentration of tropospheric ozone. They are precursors of ozone (LfU & UMEG 1999:18).

There are various regulations on the national and international level focussing on the reduction of the concentration of nitrogen oxides (NO_2, NO or NO_x) in the atmosphere. Figure 11 indicates some of these regulations. The World Health Organization (WHO) has defined NO_2 guideline levels for the protection of human health. There is a 1-hourly average guideline level of 200 µg/m³ for the assessment of short-time effects and a annual average NO_2 guideline level of 40 µg/m³ for the assessment of the long-term effects (WHO 2000). The target values of the 1st Daughter Directive (1999/30/EC) to the Air Quality Framework Directive (96/62/EC) correspond to these WHO guideline levels. In addition, within the 1st Daughter Directive a NO_x target value for the protection of vegetation from long-term effects of high concentrations of nitrogen oxides is lay down (annual average of 30 µg/m³). The target values are to be attained in 2010, this means in 2010 within the geographical scope of the EU there should be not any exceedance of the annual average NO_x concentration of 30 µg/m³ or the annual average NO_2 concentration of 40 µg/m³. For the 1-hourly average concentration of NO_2 (200 µg/m³) 18 exceedances per year are acceptable. The governments of the signatory states of the 1st Daughter Directive commit themselves to put these regulations into national legislation. The 1st Daughter Directive will replace the existing rules of the EU Directive 85/203/EEC of 7 March 1985 and on the national level the regulations of the 22.BImSchV of 26 October 1993. Under the UN ECE Convention on Long-Range Transboundary Air Pollution a effect based NO_x Critical Level for the protection of vegetation (annual average of 30 µg/m³) has been defined („Gothenburg-Protocol"). On the national level, in addition, there are the regulations of the TA Luft and the 23.BImSchV of 16 December 1996.

regulation	component	value	unit	averaging time	aspect	character	comments
WHO (1987)	NO_2	400	µg/m³	0.5-h average	human health	guideline value	
		150		24-h average			
WHO (1999)	NO_2	200	µg/m³	1-h average	human health	guideline value	
		40		annual average			
1. Daughter Directive (1999/30/EC) to Air Quality Framework Directive (96/62/EEC)	NO_2	200	µg/m³	1-h average	human health	threshold value	target value to be reached in 2010: no more than 18 exceedances of 200 µg/m³ NO_2 per year
	NO_2	40	µg/m³	annual average			target value to be reached in 2010: no exceedance permitted
	NO_X	30	µg/m³	annual average	vegetation	threshold value	target value to be reached in 2010: no exceedance permitted
UN ECE "Gothenburg protocol"	NO_X	30	µg/m³	annual average	all receptors	critical level	
22. BImSchV (85/203/EEC)	NO_2	200	µg/m³	98 percentile of the cumulative frequency of the 1-h averages of one year	human health and environment	threshold value	
23. BImSchV	NO_2	160	µg/m³	98 percentile of the cumulative frequency of the 1-h averages of one year	human health	test value	if exceeded, there should be limitations for traffic
TA Luft (1. BImSchV)	NO_2	80	µg/m³	annual average	human health	threshold value	IW1 = long-term effects
		200		98 percentile of the cumulative frequency of the 0.5-h averages of one year			IW2 = short-term effects

Figure 11: Air quality standards for the concentration of nitrogen oxides (NO_x, NO_2).

The numerous regulations had effects on the emissions of nitrogen oxides in Germany. Figure 12 indicates the development of the emissions of nitrogen oxides in Germany since 1980. Between 1980 and 1987 the emission of nitrogen oxides has been stable within Germany. The emission rates added up to about 3300 kilotons NO_2 per year. Since the end of the 1980ies there is a reduction in emission rates detectable. In 1998 nitrogen oxides emissions amounted to 1780 kilotons NO_2 – half as much as in the middle of the 1980ies. In figure 13 there is some extra information about the development of the emission rates of nitrogen oxides in the old and newly-formed German States since 1985. In the newly-formed German States the emission rate of nitrogen oxides has been stable since 1975 (about 700 kilotons/year). Since the end of the 1980ies there is a reduction detectable to 360 kilotons NO_2 in 1998. From 1985 til 1988 the emission rates of nitrogen oxides in the old West German States amounted to circa 2600 kilotons per year. Since the end of the 1980ies the emission rates are decreasing. In 1998 there has been a NO_2 emission rate of 1420 kilotons.

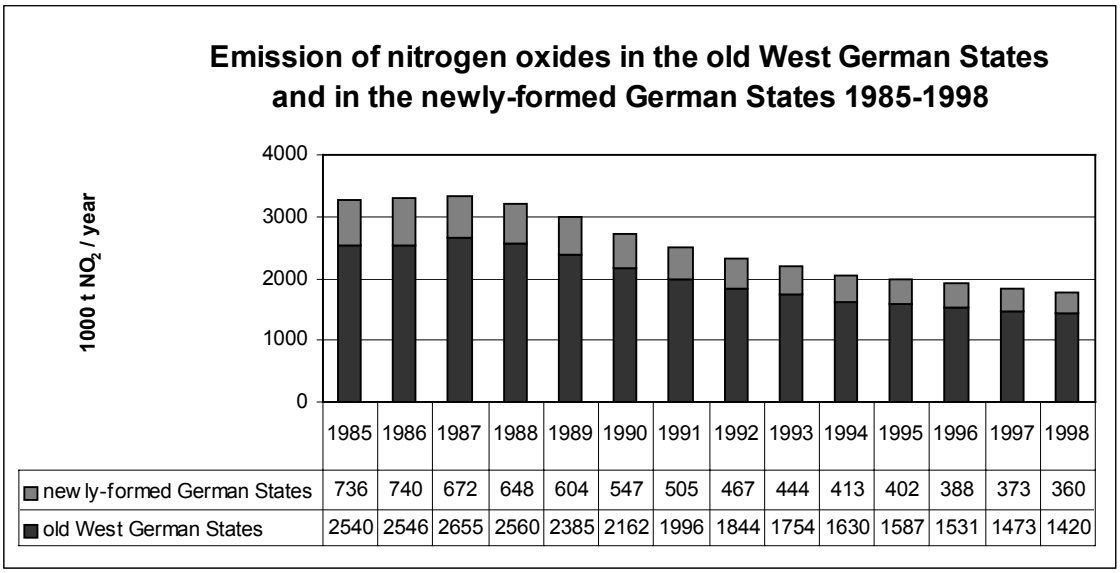


	1980	1981	1982	1983	1984	1985	1986	1987	1988	1989	1990	1991	1992	1993	1994	1995	1996	1997	1998
Emission in 1000 t NO2	3334	3259	3219	3258	3305	3276	3286	3327	3208	2989	2709	2501	2311	2198	2043	1989	1919	1846	1780

Figure 12: Emission of nitrogen oxides – expressed as 1000 t NO_2/year - in Germany 1980-1998.

The reduction of the emission of nitrogen oxides in Germany mainly is a result of the Großfeuerungsanlagenverordnung (13. BImSchV; 22 June 1983). Industrial enterprises adapted to new firings using fuels with better emission properties. Nowadays the most important emission source of nitrogen oxides are traffic exhaust gases (more than 50 %) (HELBIG et al 1999:171). In spite of technical advances in purification of waste gases (e.g. catalyst), traffic exhaust gas emissions did not diminish. The volume of traffic increased and thus compensated the effects of technical advances (HELBIG et al 1999:171). Further propagation of catalysts and the imple

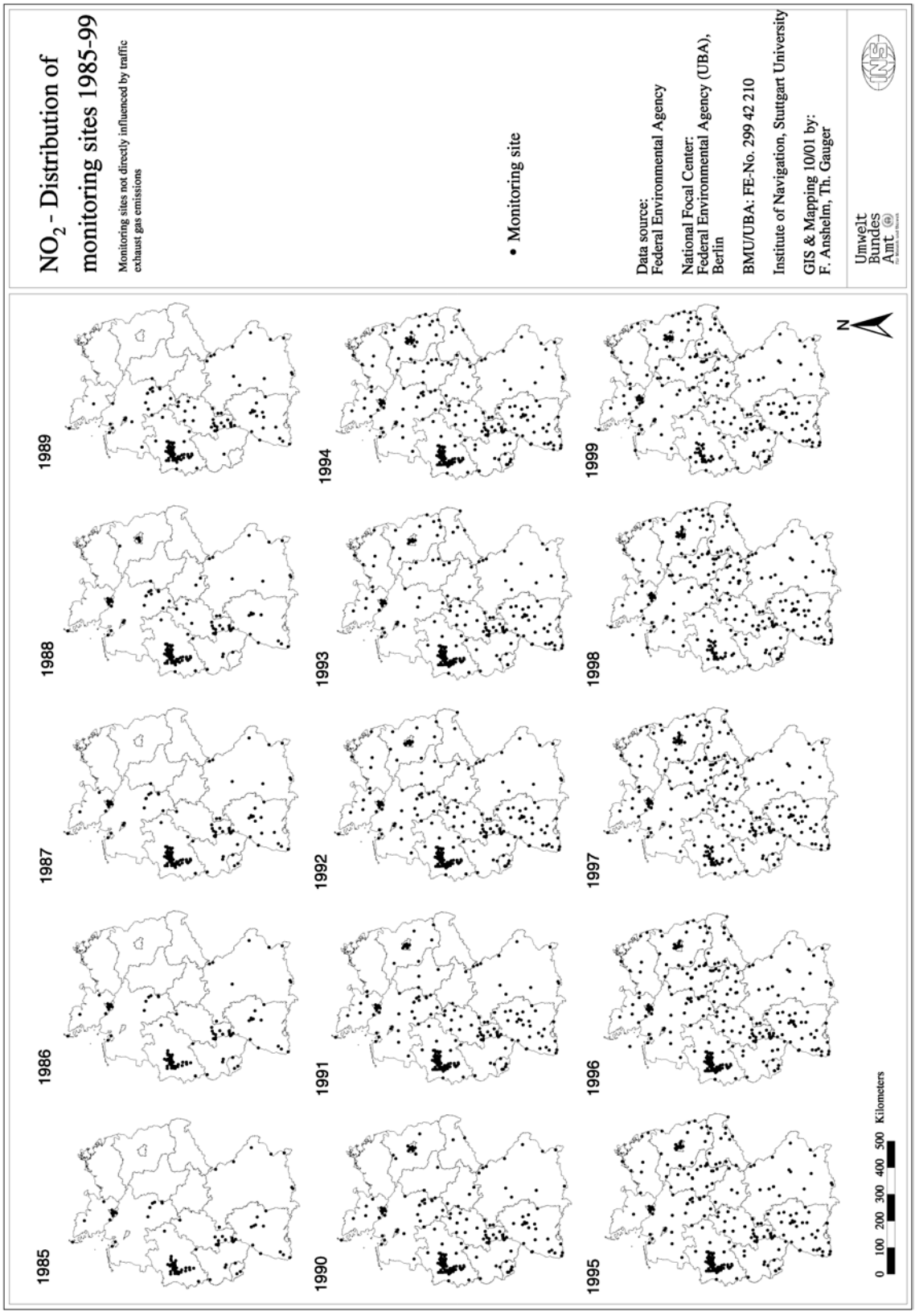
mentation of European standards concerning trucks (EURO-1- und EURO-2-engines) should cause a considerable reduction of nitrogen oxides emissions within the next years.



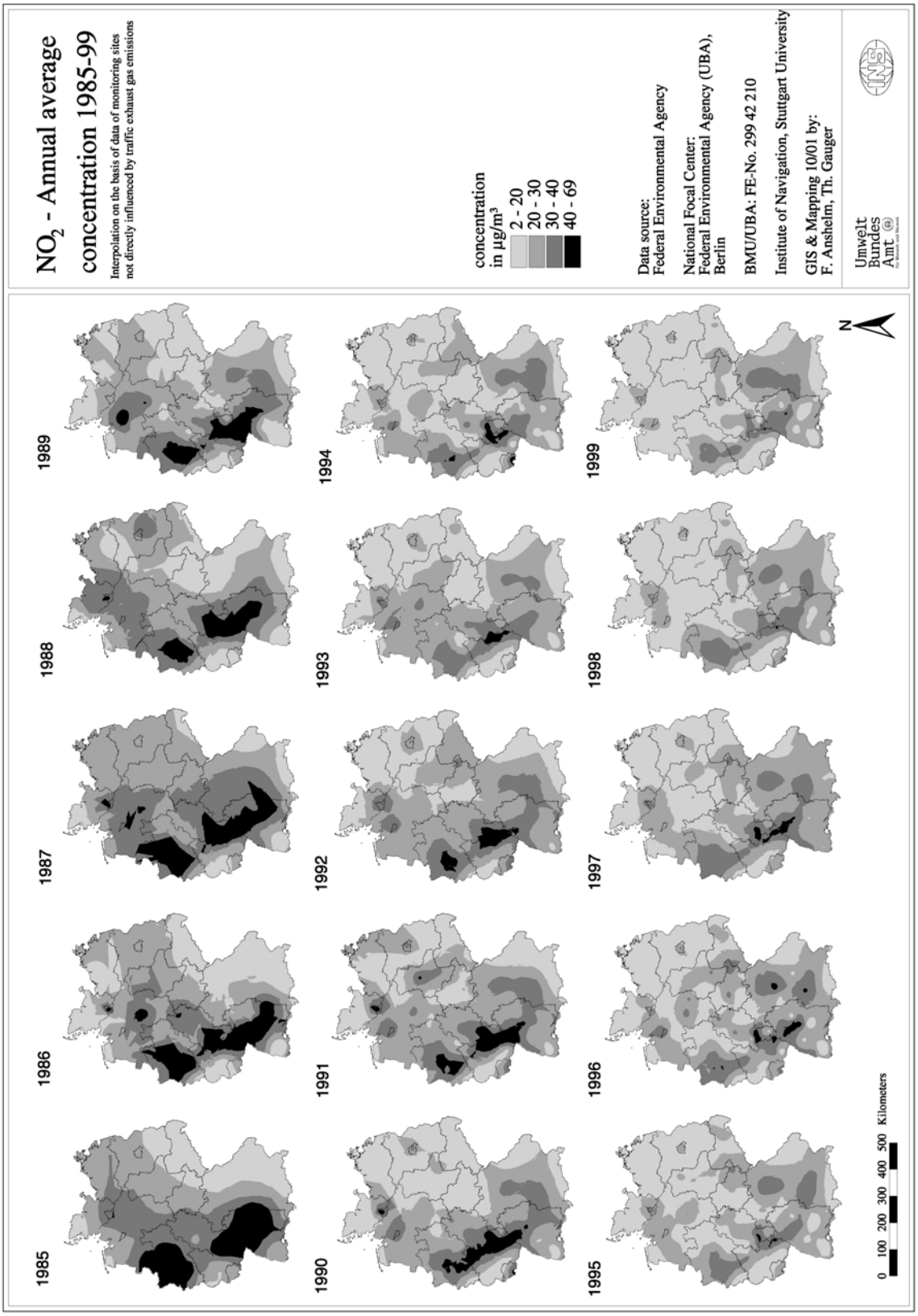
	1985	1986	1987	1988	1989	1990	1991	1992	1993	1994	1995	1996	1997	1998
newly-formed German States	736	740	672	648	604	547	505	467	444	413	402	388	373	360
old West German States	2540	2546	2655	2560	2385	2162	1996	1844	1754	1630	1587	1531	1473	1420

Figure 13: Emission of nitrogen oxides – expressed as 1000 t NO_2/year - in old West German States and newly-formed German States 1985-1998.

NO_2 concentration data are available for 1985–1999 (table 4 and figure 4). Data of up to 458 monitoring sites are stored in INS air pollutant concentrations database, but only the data of up to 275 monitoring sites are suitable for mapping NO_2 concentration (table 4). Data of monitoring sites directly influenced by traffic exhaust gas emissions are not representative for a bigger radius. Therefore only the data of monitoring sites not directly influenced by traffic exhaust gas emissions - according to the results of INS classification scheme – have been used for mapping. Mapping in a 1 · 1 km² grid has been possible for the years 1986-1999. In 1985 the set of data is too patchy to map in a small grid size (map 3). Therefore 1985 has been interpolated in a 5 · 5 km² grid (map 4). In the former German Democratic Republic region data on concentration of NO_2 strictly speaking is inadequate for mapping til 1989. Since 1990 the density of NO_2 concentration data is high enough throughout Germany.

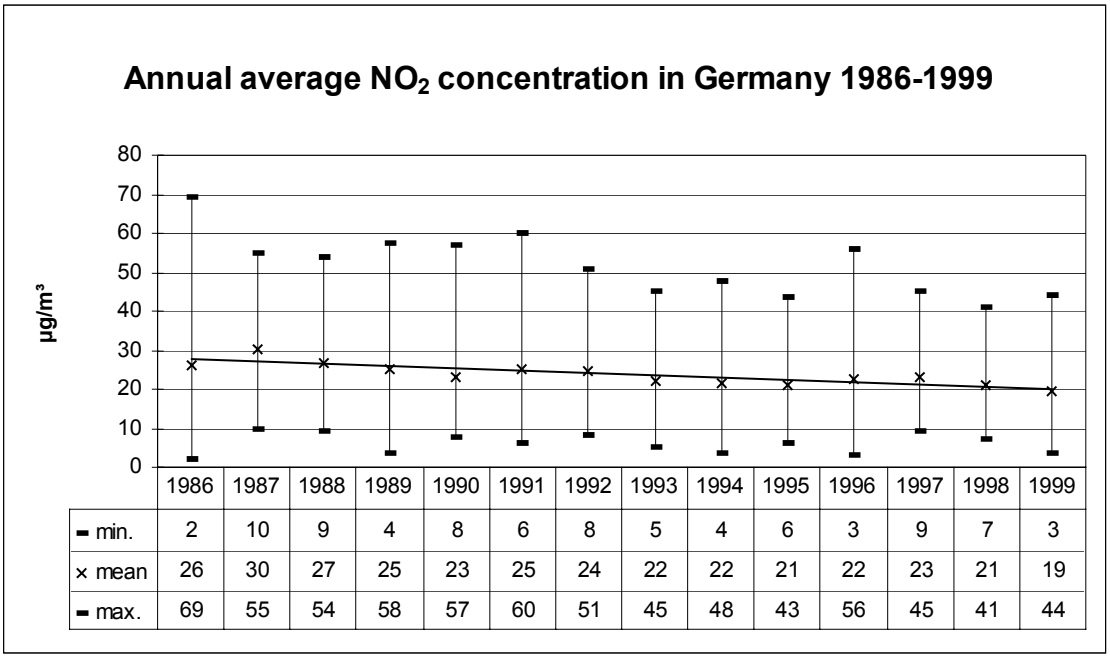


Map 3: NO_2 – Distribution of monitoring sites 1985-99.



Map 4: NO_2 – annual average concentration 1985-99.

After interpolation of the monitoring data of all monitoring sites not directly influenced by traffic exhaust gas emissions there is a slight downward trend in annual average NO_2 concentration data detectable (figure 14). The annual average concentration of NO_2 throughout Germany has been between 30 µg/m³ (1987) and 19 µg/m³ (1999). Within the single years there is, of course, a wide range of annual average NO_2 concentrations throughout Germany. In 1986 the annual average NO_2 concentration within Germany varied from 2 µg/m³ (Garmisch-Partenkirchen) to 69 µg/m³ (Mannheim/Ludwigshafen). In 1999 the annual average NO_2 concentration varies between 3 µg/m³ (Garmisch-Partenkirchen) and 44 µg/m³ (Mannheim/Ludwigshafen, Stuttgart). The range of annual average NO_2 concentrations within the single years is slowly getting smaller. Analysis of the data of the UBA monitoring network (background monitoring) indicates a slight decrease of the annual average concentration of NO_2 throughout Germany (UBA 1999: 33).



	1986	1987	1988	1989	1990	1991	1992	1993	1994	1995	1996	1997	1998	1999
– min.	2	10	9	4	8	6	8	5	4	6	3	9	7	3
× mean	26	30	27	25	23	25	24	22	22	21	22	23	21	19
– max.	69	55	54	58	57	60	51	45	48	43	56	45	41	44

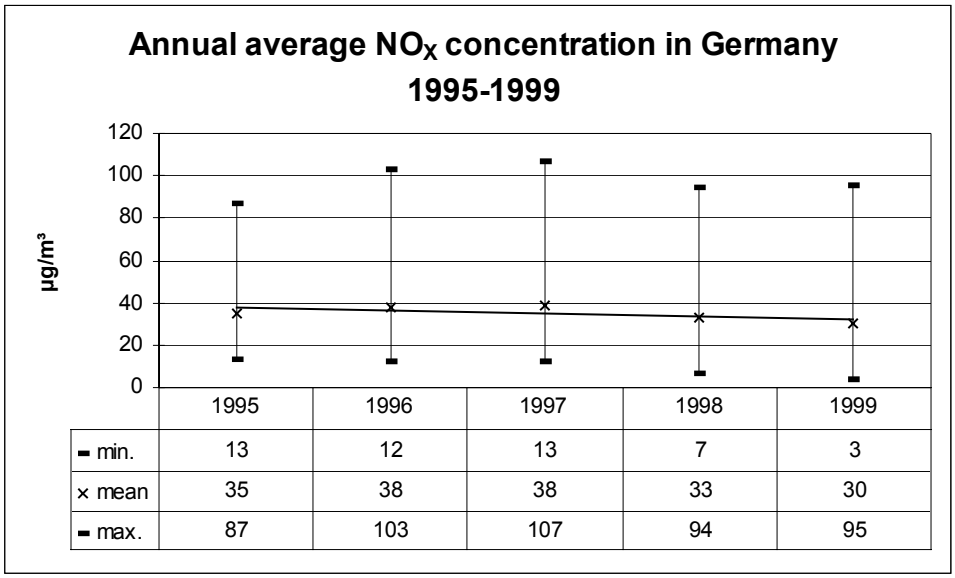
Figure 14: Annual average NO_2 concentration in Germany 1986-1999.

Regarding the spatial distribution of regions with high/low NO_2 concentrations there are some regularities visible (map 4). The Rhine region between Ruhr Basin and Rhine-Main area regularly shows relatively high NO_2 concentrations. In most of the years the annual average concentration of NO_2 is between 40 and 60 µg/m³. In some years several cities show strikingly higher NO_2 concentrations than their hinterland, e.g. Berlin in 1994 or Hamburg in 1991 (map 4).

Mecklenburg-Western Pomerania, Brandenburg, Schleswig-Holstein, the eastern parts of Bavaria and the alpine region show in most of the years relatively low NO_2 concentrations.

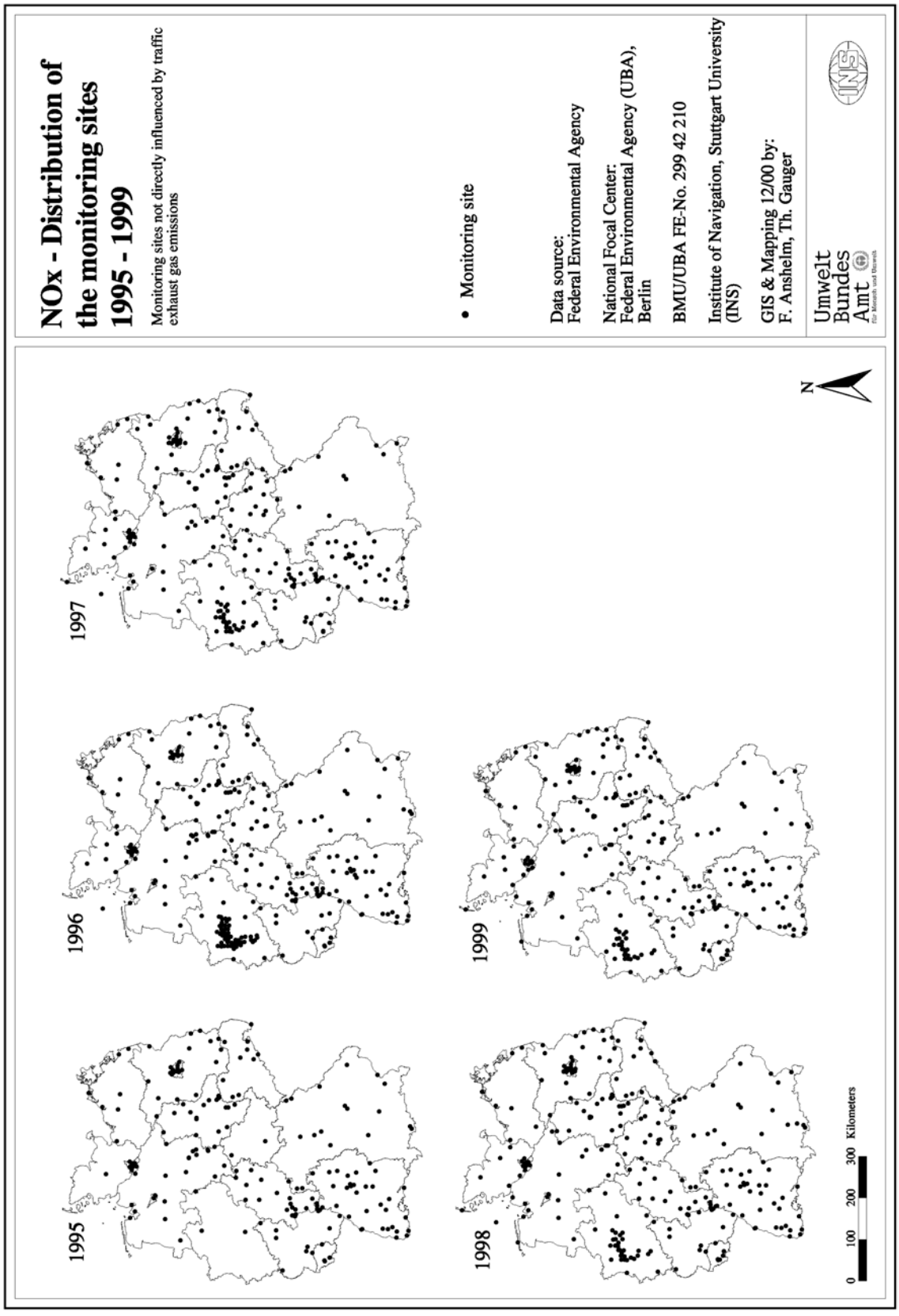
Data on the concentration of NO is available for up to 463 (1996) monitoring sites, but only the data of up to 274 (1996) NO monitoring sites is suitable for mapping (figure 4 and table 4). The series of NO monitoring data stored in INS air pollutant concentrations database starts in 1992, but before 1995 the data basis is insufficient for mapping purposes. Mapping of the concentration of NO_x therefore is only possible for the years 1995-1999 (map 5 and 6).

The NO_x concentration maps do not differ that much from the NO_2 concentration maps (map 4 and 6). The Berlin region shows higher concentration levels than the surrounding parts of Brandenburg. Highest concentration levels can be stated along the river Rhine between Ruhr Basin and Rhine-Main area. Low concentrations of NO_x can be stated in Schleswig-Holstein, except the hinterland of the city of Hamburg, in Mecklenburg-Western Pomerania, Brandenburg, the eastern parts of Bavaria and the alpine region (map 6). The series of data is too short to recognise a reliable trend (figure 15). The annual average concentration of NO_x throughout Germany has been between 30 µg/m³ (1999) and 38 µg/m³ (1996, 1997). There is no trend detectable. HELBIG et al (1999: 211) recognise at least for the 1990ies a slight decreasing trend in annual average concentration of NO_x. Til 1990 HELBIG et al (1999: 211) state a slight increase of the annual average concentration of NO_x as well in rural as in urban areas.

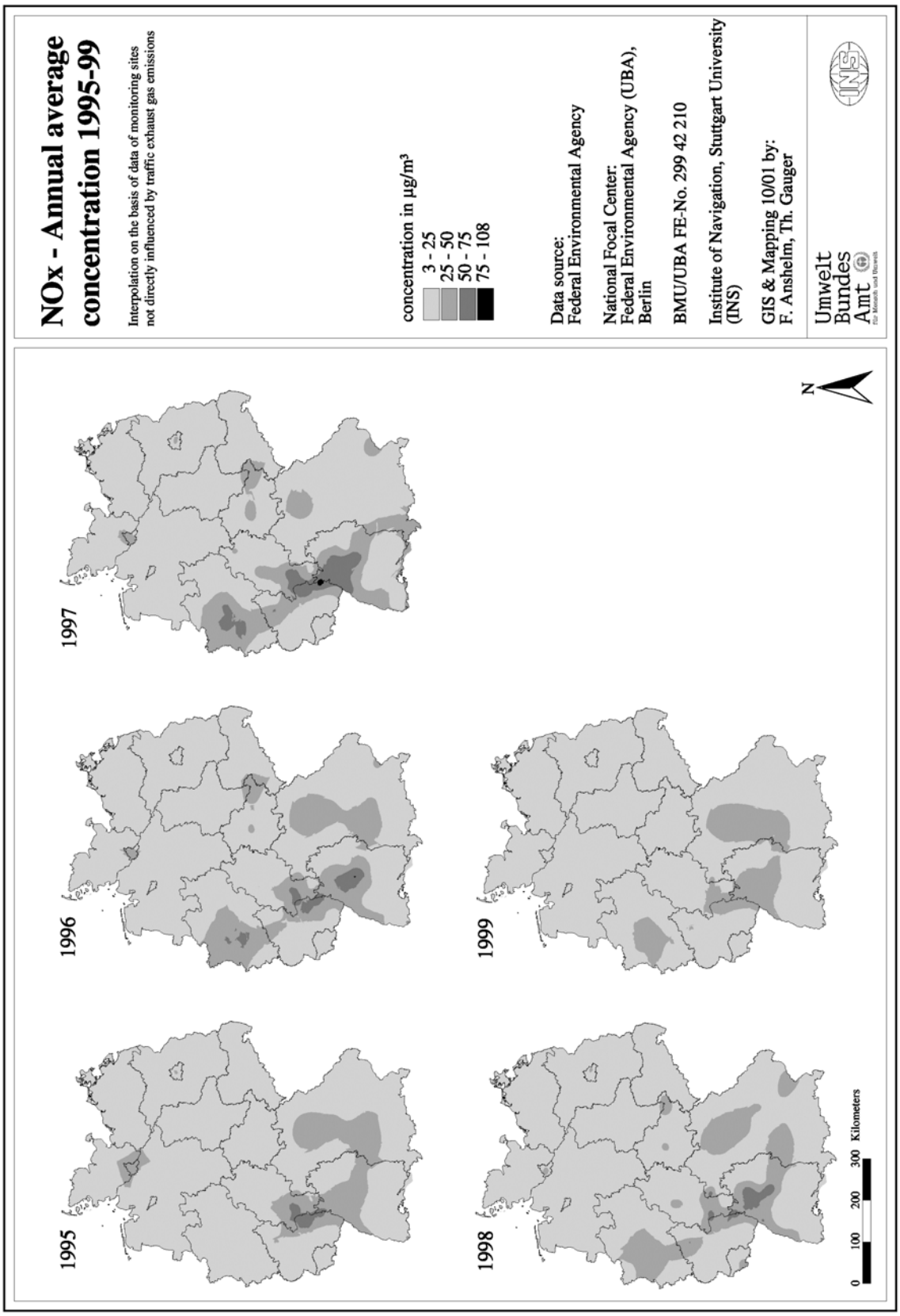


	1995	1996	1997	1998	1999
▬ min.	13	12	13	7	3
× mean	35	38	38	33	30
▬ max.	87	103	107	94	95

Figure 15: Annual average NO_X concentration in Germany 1995-1999.



Map 5: NO_x – Distribution of monitoring sites 1995-99.



Map 6: NO_x – annual average concentration 1995-99.

The mapping results show strikingly differences between the old West German States and the newly-formed German States. The annual average NO_x concentration in the newly-formed German States is explicitely lower than in the old West German States. Between 1995 and 1999 the annual average NO_x concentration in the newly-formed German States has been between 34 µg/m³ (1999) and 43 µg/m³. In the old West German States the annual average NO_x concentration has been between 22 µg/m³ (1999) and 29 µg/m³ (1996). The series of annual data is too short (1995-1999) to recognise reliable trends. In fact, there is no clear trend visible between 1995 and 1999. When looking at the longer time series of NO_2 concentration data there is a slight downward trend in the newly-formed German States as well as in the old West German States detectable. The annual average and maximum NO_2 concentration is strikingly lower in the newly-formed German States (about 20-30%). In 1991 the annual average NO_2 concentration in the newly-formed German States has been higher than in the years before, maybe because of a increase in the volume of traffic after the German Unification. But looking at the emission data this aspect can not be detected (figure 12).

3.3 Mapping concentration of O_3

Tropospherical ozone is produced through photochemical processes. It mainly results from the sunlight-initiated oxidation of volatile organic compounds (VOCs) in the presence of nitrogen oxides (NO_x = NO and NO_2). Nitrogen oxides and VOCs are precursor compounds for ozone production in the troposphere. Besides, CH_4 and CO are important precursor compounds. Nitrogen oxides as well as VOCs are emitted by natural processes, in soils and vegetation respectively. However, the anthropogenic emissions of these gases through combustion, especially by motor vehicles and industrial processes, exceed the natural emissions by far.

Elevated concentrations of ozone have become a problem, because ozone is phytotoxic, has a damaging effect on certain materials (rubber products, plastics, paints) and last but not least represents a potential threat to human health[4]. There are numerous regulations on the national and international level concerning ground-level ozone concentrations and their effects. Figure 16 gives a review of some of these regulations. In 1987 there has been a recommendation of the World Health Organization (WHO 1987) concerning ground-level ozone. The World Health Organization suggests a value of 150-200 µg/m³ for maximum short-time concentration of ozone and a value of 100-120 µg/m³ as a 24-hours mean value as a guideline. Between 1993 and

[4] topic ozone and health effects will be discussed into detail in chapter 5.

regulation	component	value	unit	averaging time	aspect	character	comments
WHO (1999)	O_3	120	µg/m³	8-h movingaverage	human health	guideline value	8-h moving average of one year
3. Daughter Directive (proposal) to Air Quality Framework Directive (96/62/EC)	O_3	120	µg/m³	8-h moving average	human health	target value	8-h moving average of one year; no more than 20 days per calendar year averaged over 3 years; target year 2010
		180	µg/m³	1-h average		information value	information of the population
		240				alert value	threshold for setting off ozone alarm system
		17000	µg/m³	AOT40	vegetation	target value	May-July;8.00-20.00; averaged over 5 years; target year 2010
		6000				long-term objective	8-h moving average ; no exceedance
UN ECE "Gothenburg protocol"	O_3	6000	µg/m³	AOT40	crops and natural vegetation	critical level	May-July; 8.00-20.00
		20000			forests		April-September; 8.00-20.00
		0		AOT60	human health		January-December; 0.00-24.00; calculated as a surrogate for the WHO Air Quality Guideline (WHO 1999)
22.BImSchV (92/72/EEC)	O_3	110	µg/m³	8-h average	human health	threshold value	0.00-8.00; 8.00-16.00; 16.00-24.00; 12.00-20.00
		180		1-h average		information value	information of the population
		360		1-h average		alert value	threshold for setting off ozone alarm system
		65		24-h average	vegetation	threshold value	
		200		1-h average			

Figure 16: Air quality standards for ozone.

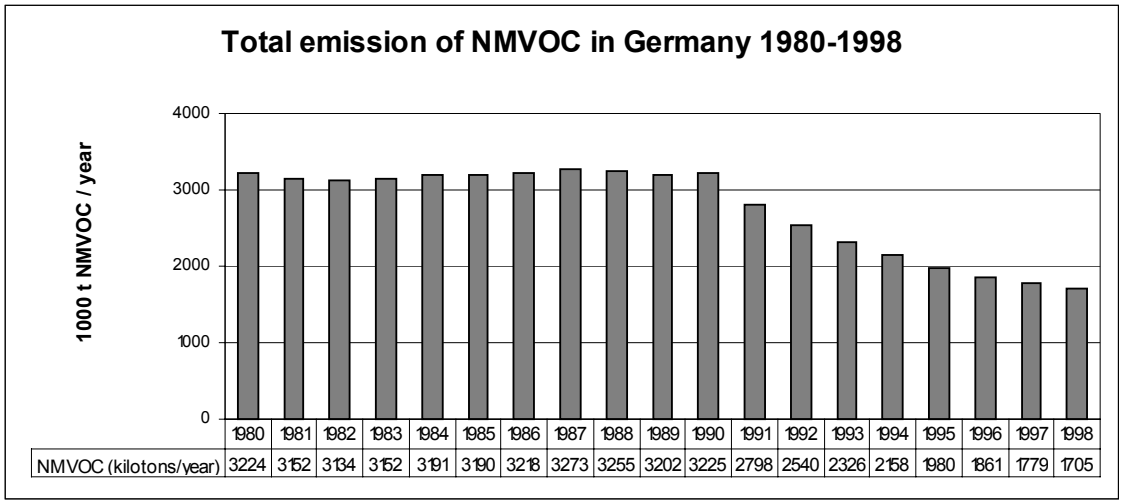
1996 the WHO Regional Office for Europe revised the 1987 guidelines. Latest findings in medicine and ecotoxicology have been considered. The old guidelines (1-hourly maximum of 150-200 µg/m³ and 24-h mean value of 100-120 µg/m³) will be abolished as soon as the new guidelines will come into force. This is expected to be in 2001. There will be a hourly running 8-hours average value of 120 µg/m³ as a guideline for ozone concentration in the revised WHO Air Quality Guidelines for Europe.

Within the framework of the UN/ECE Convention on Long-Range Transboundary Air Pollution (UN/ECE CLRTAP) the ozone AOT40 exposure index has been developed. The UN/ECE Protocol to Abate Acidification, Eutrophication and Ground-Level Ozone („Gothenburg Protocol“) defines Critical Levels for the protection of crops and natural vegetation (AOT40 of 6000 µg/m³), forests (AOT40 of 20000 µg/m³) and human health (AOT60 of 0 µg/m³). The Executive Body adopted the Protocol to Abate Acidification, Eutrophication and Ground-level Ozone in Gothenburg (Sweden) on 30 November 1999. Sixteen ratifications are needed for the Protocol to come into force. Presently there is only one ratification (September 2001).

Within the framework of the EC Air Quality Framework Directive (96/62/EC) further regulations relating to ground-level ozone concentrations have been adopted, for instance the current Ozone Directive (92/72/EEC). On 9 June 1999 the Commission's proposal for the 3rd Daughter Directive on ozone appeared. This Directive eventually will replace the current Ozone Directive (92/72/EEC). The proposal for the 3rd Daughter Directive sets long-term objectives equivalent to the World Health Organisation's new guideline levels and the UN ECE protocol's Critical Levels. Interim target values are set for ozone in ambient air to be achieved as far as possible within a given time frame (until 2010). To protect human health an 8-hours moving average of 120 µg/m³ should not be exceeded (long-term objective). Till 2010 there should be no more than 25 days per calendar year - averaged over three years – with exceedance of the 8-hours average of 120 µg/m³. To protect vegetation from unswallowable damages caused by high ozone concentrations an AOT40 of 6000 µg/m³ (long-term objective) or 17000 µg/m³ (target value for the year 2010) respectively should not be exceeded. Non-compliance requires Member States to work out reduction plans and programmes to be reported to the Commission and to be made available to the public so as to allow citizens to trace progress towards meeting the ozone standards. The proposal includes also improved and more detailed requirements to monitor and assess ozone concentrations and to inform citizens about the actual pollution load. The requirements of the 3rd Daughter Directive to the Air Quality Framework Directve (96/62/EC) will be put into national legislation like it has been done with the Ozone Directive (92/72/EEC →

22.BImSchV). The 22.BImSchV and the Ozone Directive (92/72/EEC) already contain threshold values for the protection of human health, vegetation and in addition an information value (1-h average of 180 µg/m³) for the information of the population on high ozone concentrations and an alert value of 360 µg/m³ (1-h average) for setting off ozone alert system.

In order to fulfil the requirements of the various regulations emission abatement strategies have been developed. Figure 17 and 18 illustrate the development of the emission rates of NMVOC (non-methane volatile organic compounds) in Germany. The development of the emission rates of NO_x have already been described in chapter 3.2. The emission rates of NMVOC have been almost stable between 1980 and 1990 (about 3200 kilotons/year). Since 1991 the emission rate decreases continuously. In 1998 it amounted to just 1705 kilotons. Figure 18 illustrates differences in emission rates existing between the old West German States and the newly-formed German States. In the newly-formed German States emission rates amounted to 743 kilotons in 1985. From 1986 (762 kilotons) to 1998 (273 kilotons) emission rates decreased continuously. In the old West German States emission rates amounted to 2447 kilotons in 1985. From 1987 (2749 kilotons) to 1998 (1432 kilotons) emission rates decreased continuously.



	1980	1981	1982	1983	1984	1985	1986	1987	1988	1989	1990	1991	1992	1993	1994	1995	1996	1997	1998
NMVOC (kilotons/year)	3224	3152	3134	3152	3191	3190	3218	3273	3255	3202	3225	2798	2540	2326	2158	1980	1861	1779	1705

Figure 17: Total emission of NMVOC in Gemany 1980-1998 (EMEP/MSCW 2000).

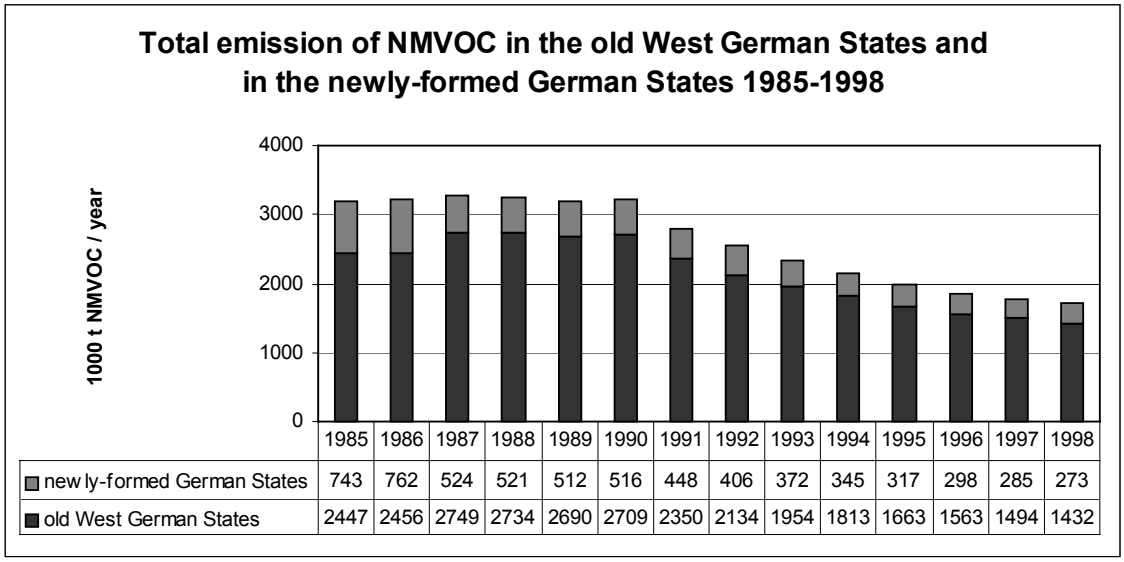


Figure 18: Total emission of NMVOC in the old West German States and newly-formed German States 1985-1998 (EMEP/MSCW 2000).

Ozone concentration data are available for 1988–1999, but mapping is only possible for the years 1990-1999. The data of the years 1988 and 1989 are too patchy for mapping purposes. The series of ozone monitoring data suitable for mapping purposes starts in 1990. In 1990 there are 110 monitoring sites, which are not directly influenced by traffic exhaust gas emissions (table 4). Till the end of the 1990ies the number of monitoring sites suitable for mapping increased up to more than 210. Map 7 illustrates the distribution of monitoring sites suitable for mapping. In the early 1990ies there still have been some bigger gaps in the monitoring network, especially in the newly-formed German States, Lower Saxony and Bavaria. Annual average ozone concentration data from monitoring sites not directly influenced by traffic exhaust gas emissions have been interpolated by using Kriging technique. The reliability of the interpolated data strongly depends on the quality of the monitoring data and the density of the underlying monitoring network. A high density of monitored data favours good interpolation results. At the latest since 1995 there is a real good distribution of monitoring sites within Germany.

Map 8 illustrates mapping results for annual average ozone concentration in Germany from 1990 to 1999. There are strong spatio-temporal variations in ozone concentration. This variability is mainly due to the different weather conditions in different years and different places. But of course, regional differences in the amount of ozone precusor compounds in ambient air plays an important role, too. High temperatures at the same time with stagnant high-pressure weather systems (high radiation) with low wind speeds (reduced exchange of polluted air masses) as

well as a high amount of ozone precursor compounds contribute to harmful ozone accumulation.

Regularly recurring high annual average ozone concentrations occur at remote mountain areas of high elevation (Black Forest, The Alps, Bavarian Forest, Thuringian Forest, Harz, etc). In high altitudes the concentration of NO and therefore the depletion of ozone is lower. Secondly, mountain areas are sometimes influenced by air masses of the free troposphere (mainly in winter time). And ozone concentrations are relatively high in the free troposphere. Finally, as windspeeds and turbulent mixing processes are enhanced on hills, it follows that surface depletion effects (dry deposition) are smaller in mountain areas than in sheltered lowlands. Figure 19 illustrates the dependency between annual average ozone concentration and altitude on the basis of ozone monitoring data of 1999 (sites not directly influenced by traffic exhaust gas emissions only). Highest annual average ozone concentrations can be found in remote mountain areas.

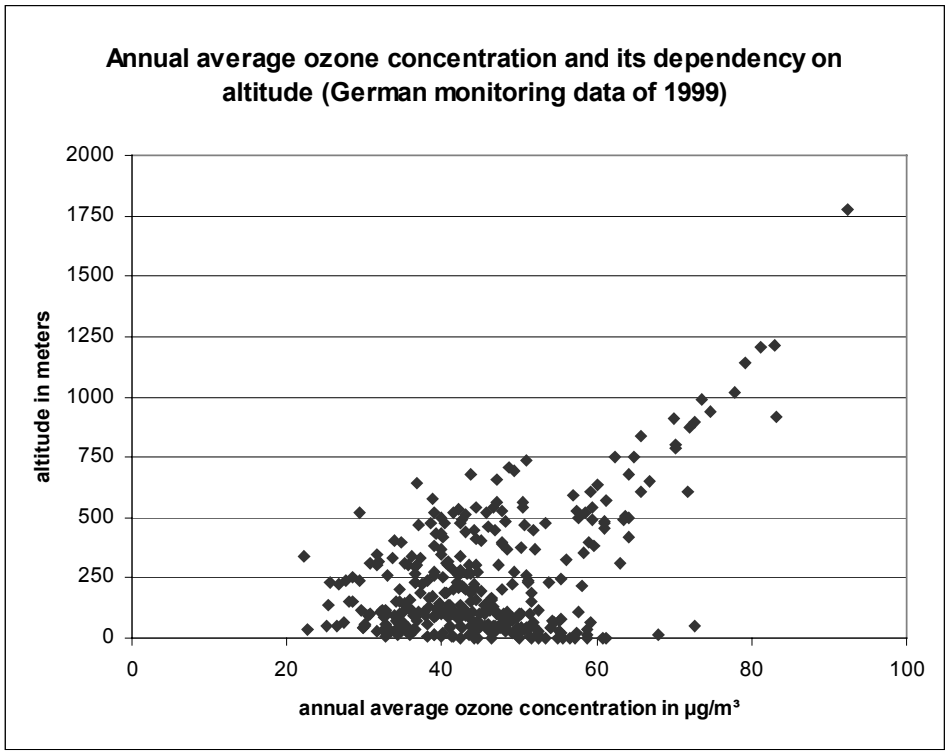


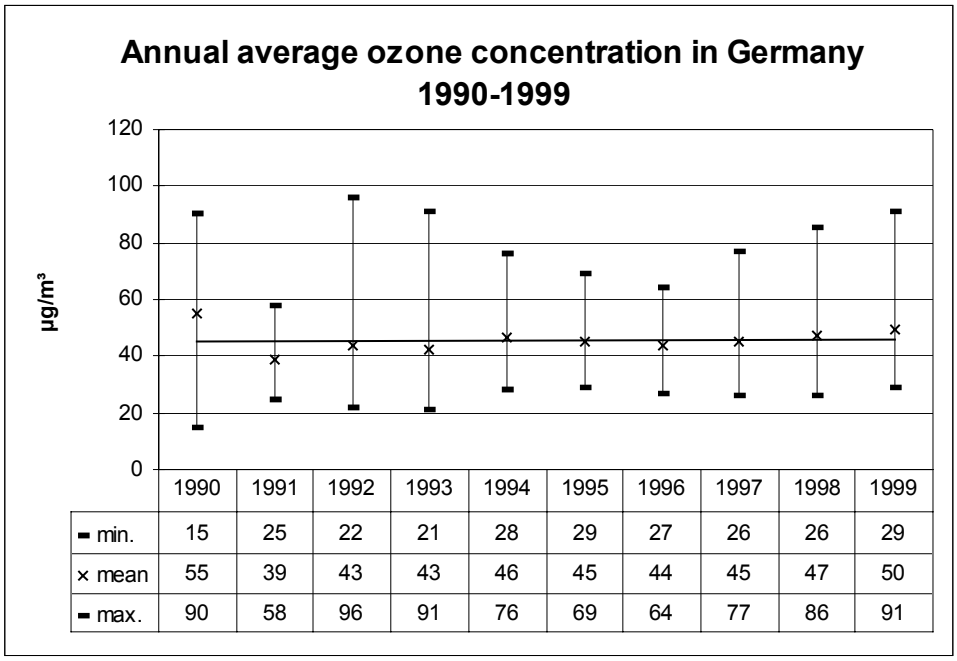
Figure 19: Ozone concentration and its dependency on altitude (German monitoring data of 1999).

Map 8 corroberates this statement. Highest annual average ozone concentrations can be found in mountain areas (Black Forest, Bavarian Forest, Erzgebirge, Harz, Thuringian Forest, The Alps). Low ozone concentrations normally can be found in urban areas due to the reaction of ozone

with nitric oxide, which generates NO_2 and depletes ozone. The main source of NO in populated areas is from vehicle exhaust gas emissions. Data of monitoring sites directly influenced by traffic exhaust gas emissions have not been used for calculating ozone concentration maps. But nevertheless there might be some effects of traffic exhaust gas emissions-initiated low urban ozone concentrations left in the mapping results. In congested urban areas like the cities of Berlin, Hamburg and Bremen or in North Rhine-Westphalia (Ruhr Basin) ozone concentrations are relatively low in most of the years. This might be an effect of the high traffic density in these regions.

In order to detect regional phenomenons despite large inter-year variabilities due to weather conditions a 10 years average ozone concentration map has been calculated (map 9). Map 9 confirms the statements made above. There are high annual average ozone concentrations at remote mountain areas (Black Forest, Bavarian Forest, Harz, The Alps) and there are low ozone concentrations in congested urban areas (Berlin, Leipzig, Hamburg, Bremen, Saarbrücken, Rhine-Main area, North Rhine-Westphalia).

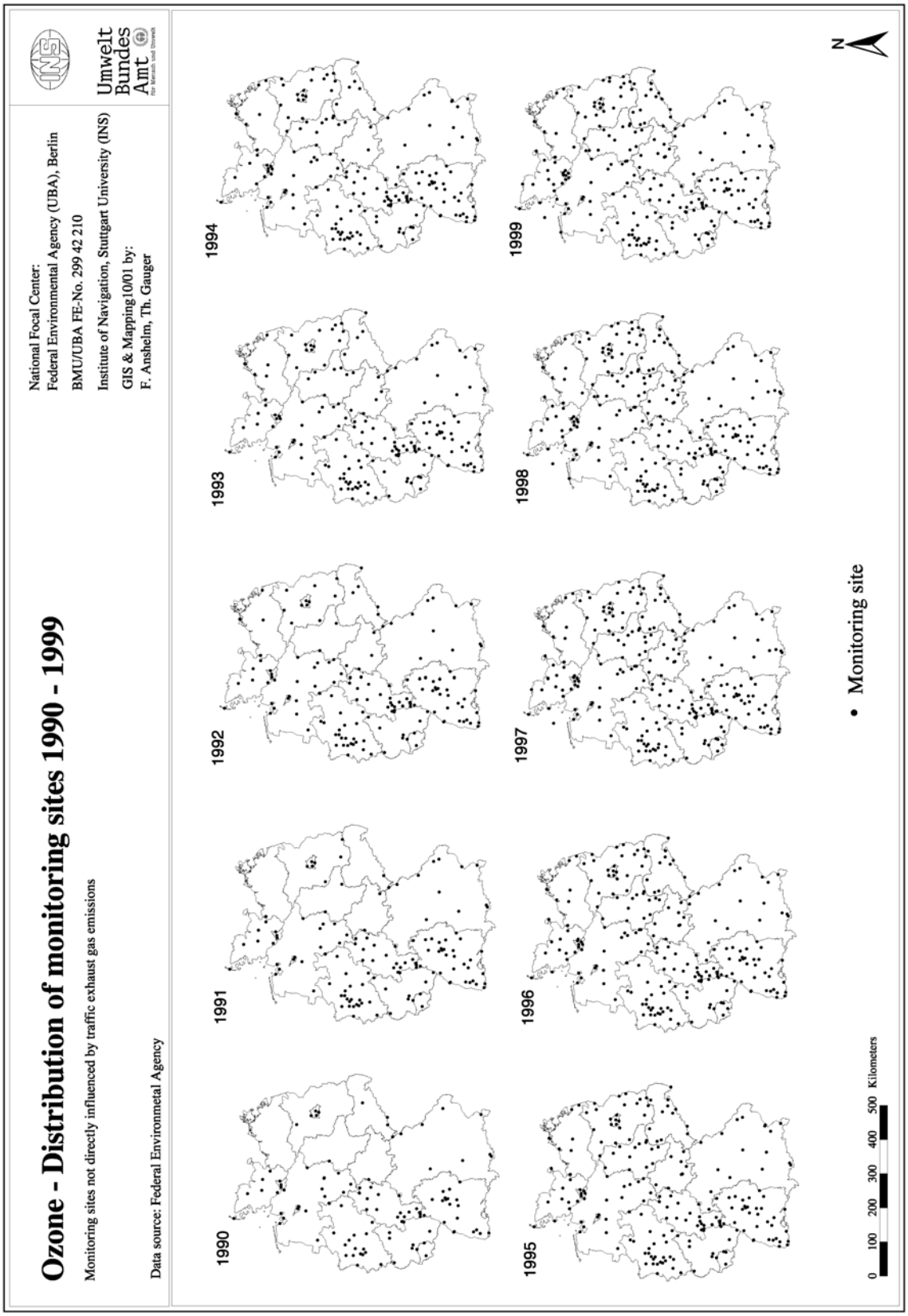
In the 1990ies there is no clear cut trend in annual average ozone concentration data in Germany (figure 20). The annual average ozone concentration varied between 39 µg/m³ (1991) and 55 µg/m³ (1990). These values are averaged over the whole area of Germany. In every single year there is, of course, a wide range of annual average ozone concentrations throughout Germany. There are areas with explicitely lower or higher annual average ozone concentrations. In 1992, for instance, there are small areas near to Hamburg and Frankfurt, where the annual average ozone concentration has amounted to not more than 23 µg/m³. In 1992 the maximum annual average ozone concentration (96 µg/m³) has been reached in the area of the Black Forest.



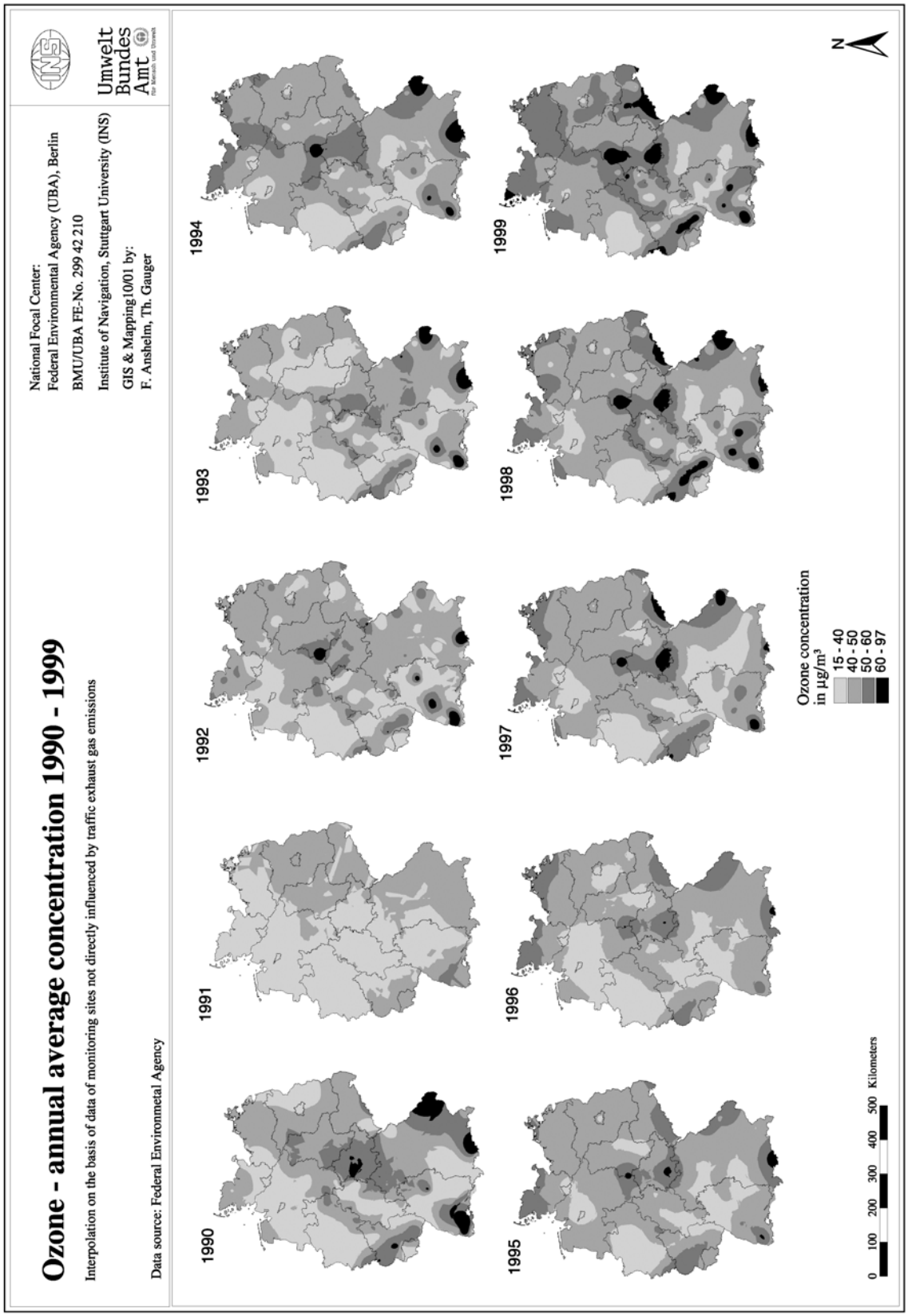
	1990	1991	1992	1993	1994	1995	1996	1997	1998	1999
▬ min.	15	25	22	21	28	29	27	26	26	29
× mean	55	39	43	43	46	45	44	45	47	50
▬ max.	90	58	96	91	76	69	64	77	86	91

Figure 20: Annual average ozone concentration in Germany 1990-1999.

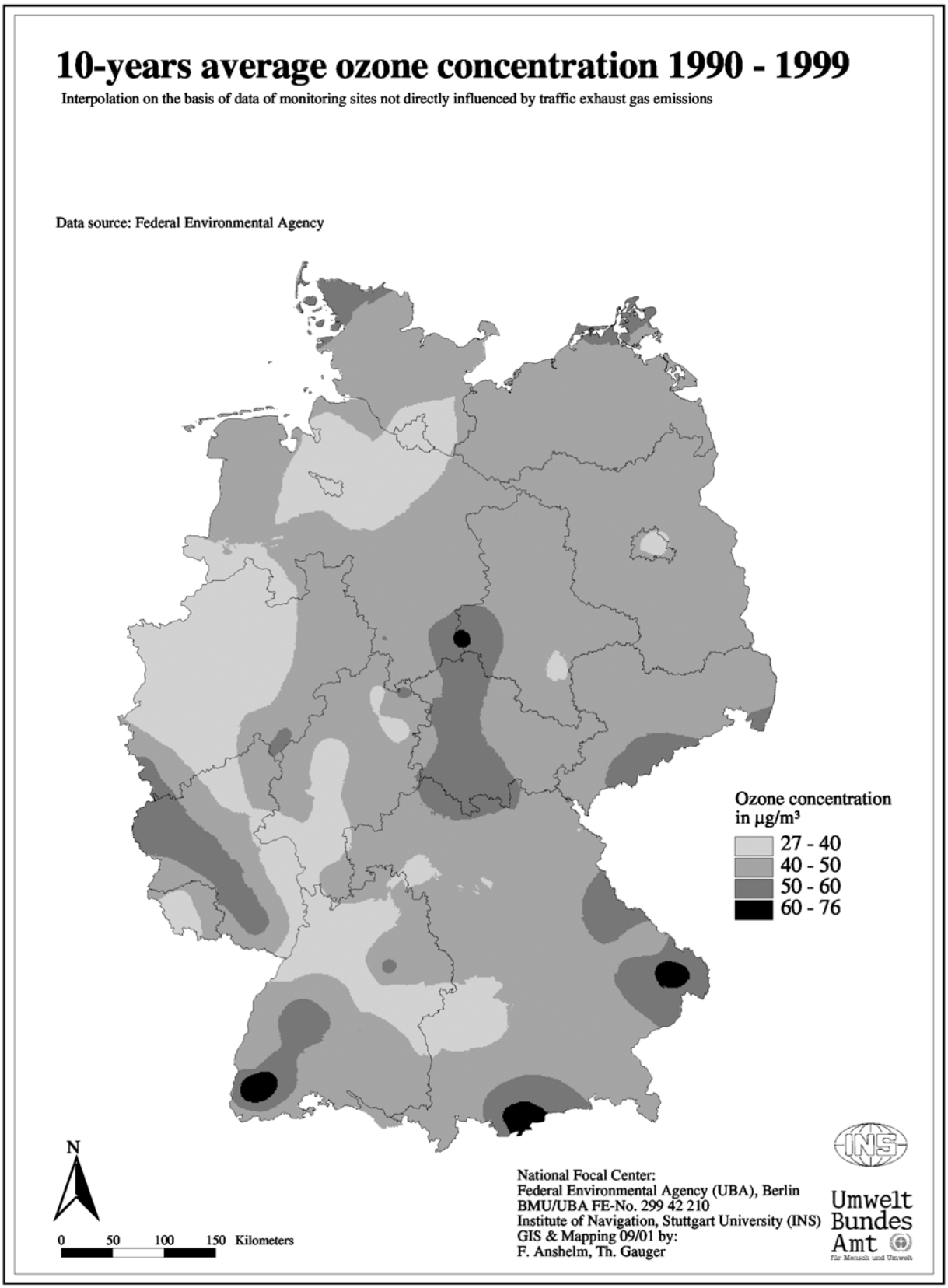
These statements refer to statistical analysis of the mapping results and not to statistical analysis of the monitored data itself. Due to the interpolation method the maxima become a little lower and the minima become a little higher.



Map 7: Ozone – Distribution of monitoring sites 1990-99.



Map 8: Ozone – annual average concentration 1990-99.


10-years average ozone concentration 1990 - 1999
Interpolation on the basis of data of monitoring sites not directly influenced by traffic exhaust gas emissions
Data source: Federal Environmental Agency
Ozone concentration in µg/m³
27 - 40
40 - 50
50 - 60
60 - 76
N
0 50 100 150 Kilometers
National Focal Center:
Federal Environmental Agency (UBA), Berlin
BMU/UBA FE-No. 299 42 210
Institute of Navigation, Stuttgart University (INS)
GIS & Mapping 09/01 by:
F. Anshelm, Th. Gauger
INS
Umwelt Bundes Amt
für Mensch und Umwelt

Map 9: 10-years average ozone concentration 1990-99.

4 Mapping Critical Levels exceedances

The **Critical Loads and Levels concept** was adopted as a policy making tool in development of emission control strategies within the United Nations Convention on Long-Range Transboundary Air Pollution (signed in 1979). **Critical Levels** are defined as concentrations of pollutants in the atmosphere above which direct adverse effects on receptors, such as human beings, plants, ecosystems or materials, may occur according to present knowledge. Critical Levels consider the receptor specific sensitivity against different kinds of corrosive gases. The sensitivity of the receptor decides on the reference period which has to be considered in risk assessment. Critical Levels exceedance maps show the difference between the mapped monitored air pollutant concentration or AOT40 data and the Critical Levels.

According to GREGOR (1993) permanent re-evaluation of the standard of knowledge is an essential part of the concept of Critical Levels & Critical Loads. As a result most of the Critical Levels have changed over the years. The decisions which lead to the definition of Critical Levels are based on scientific findings, which have been presented on the UN ECE workshops in Bad Harzburg (UN ECE 1988), Egham (UN ECE 1992), Bern (UN ECE 1993a) and Kuopio (UN ECE 1996a). Figure 21 describes the development of the Critical Levels for SO_2, NO_x, NH_3 and O_3 since 1988. The Critical Levels presently valid are marked in bold type. Only these Critical Levels will be illustrated below.

For SO_2 the Critical Levels defined on the UN ECE workshop in Egham (1992) are still valid. Lichens are most sensitive to SO_2. The Critical Level therefore amounts to an annual average of only 10 µg/m³. For forest ecosystems and natural vegetation the Critical Level is 20 µg/m³. The annual average concentration as well as the average concentration of the winter half year (October – March) should not exceed this value. Due to the particularly high sensitivity of plants which grow in high altitudes the Critical Level for forest ecosystems and natural vegetation is set to 15 µg/m³ when the annual temperature sum above 5 °C (diurnal average) amounts to less than 1000 °C-days. Crops are considered to be less sensitive to SO_2. The Critical Level for crops therefore amounts to 30 µg/m³ (annual average and average of October – March).

The Critical Levels for NO_x refer to all kinds of terrestrial plants. An annual average concentration of 30 µg/m³ should not be exceeded. In order to adress short-term effects of particularly high NO_x concentrations too, a 4-hours average concentration of 95 µg/m³ should not be exceeded.

The Critical Levels for NH_3 consider different reference periods, but do not distinguish between different kinds of receptors. The annual average concentration of NH_3 should not exceed 8 µg/m³. In addition, a monthly average of 23 µg/m³, a 24-hours average of 270 µg/m³ and a 1-hour average of 3300 µg/m³ should not be exceeded.

While the Critical Levels for SO_2, NO_x and NH_3 persist since the Egham workshop (UN ECE 1992) the Critical Levels for ozone have been modified in Bern (UN ECE 1993a) and, once more, in Kuopio (UN ECE 1996a). The Critical Levels for ozone presently in force are based on the concept of AOT40 exposure index (Accumulated exposure Over a Threshold of 40 ppb). AOT40 exposure index takes into account not only the peak values, but also the frequency of high ozone concentrations. Calculation of AOT40 exposure index is explained in chapter 4.3. There is a Critical Level of 3000 ppb.h for crops and semi-natural vegetation (AOT40crops) and a Critical Level of 10000 ppb.h for forest ecosystems (AOT40forests). AOT40crops and AOT40forests refer to different reference periods. AOT40crops is calculated on the basis of the ozone concentration data of the daylight hours of May to July, AOT40forests is calculated on the basis of the ozone concentration data of the daylight hours between April and September. In addition, short-term Critical Levels for crops have been defined.

4.1 Exceedance of the Critical Levels for SO_2

The Critical Levels for SO_2 which have been established in Egham in 1992 (UN ECE 1992) are still valid. There are Critical Levels for three different types of receptors – for lichens (10 µg/m³: annual average and average the winter half year), for forest ecosystems and natural vegetation (20 µg/m³: annual average and average of the winter half year) and for crops (30 µg/m³: annual average and average of the winter half year).

Exceedance of the Critical Levels for SO_2 has been calculated and mapped for the years 1985-1999 (map 10, 11 and 12). The exceedance maps reflect the decline in SO_2 concentration since 1985. Highest exceedances in the early years, little exceedances at the end of the 1990ies.

In the late 1980ies the SO_2 Critical Level for lichens has been exceeded up to a level of 4483 % (map 10). The Critical Level has been exceeded almost throughout Germany. Since 1988 the areas with no exceedance become bigger. In Bavaria, Baden-Württemberg and Schleswig-Holstein the exceedances have been smallest. Since 1991 the exceedances were decreasing in

Figure 21: Critical Levels for SO_2, NH_3, NO_x und O_3.

corrosive gas	critical levels[a] 1988	1992	1993	1996	reference period	receptor
SO_2 [µg/m³]	70[b]	-			maximum 24-h average	overall vegetation
		10			annual average	lichens
	20	-			annual average	forest ecosystems and natural vegetation
		20			annual average and 6 months average October - March	
		15			annual average, when the sum of daily mean temperature above 5 °C is less than 1000 °C-days	
	30	-			annual average	crops
		30			annual average and 6 months average October - March	
NO_X [µg/m³]		**95**			maximum 4-h average	overall vegetation
	30	**30**			annual average	
NH_3 [µg/m³]	1000	**3300**			maximum 1-h average	overall vegetation
	600	**270**			maximum 24-h average	
	100	**23**			maximum monthly average	
		8			annual average	
O_3 [µg/m³]	150	-			maximum 1-h average	overall vegetation
	60	-			maximum 8-h average between 1a.m. - 8 a.m., 9 a.m. - 4 p.m. and 5 p.m. - 12 p.m.	
	50	-			maximum 7-h average; 9 a.m. - 4 p.m. during vegetation period (possibly April - September)	
O_3 AOT40[c] [ppb.h]		300	-		vegetation period; daylight hours; between astronomic sunrise and sunset	overall vegetation
			5300	**3000**	3 months of the main growing season (May - July); daylight hours[d]	crops and semi-natural vegetation
			700	-	3 days running; only daylight hours	crops
				500	5 days running; daylight hours with vapor pressure deficit > 1.5 kPa (average from 9.30 a.m. - 4.30 p.m.)	
				200	5 days running; daylight hours with vapor pressure deficit < 1.5 kPa (average from 9.30 a.m. - 4.30 p.m.)	
			10000	-	6 months of highest sensitivity of the receptor (April - September); full-time	forest ecosystems
				10000	6 months of highest sensitivity of the receptor (April - September); daylight hours	

[a] critical levels have been established on the basis of results of the UN ECE workshops in Bad Harzburg (UN ECE 1988), Egham (UN ECE 1992), Bern (UN ECE 1993) and Kuopio (UN ECE 1996)

[b] critical levels in cursive types mean that the critical level has been revised, the critical levels in bold type are still valid

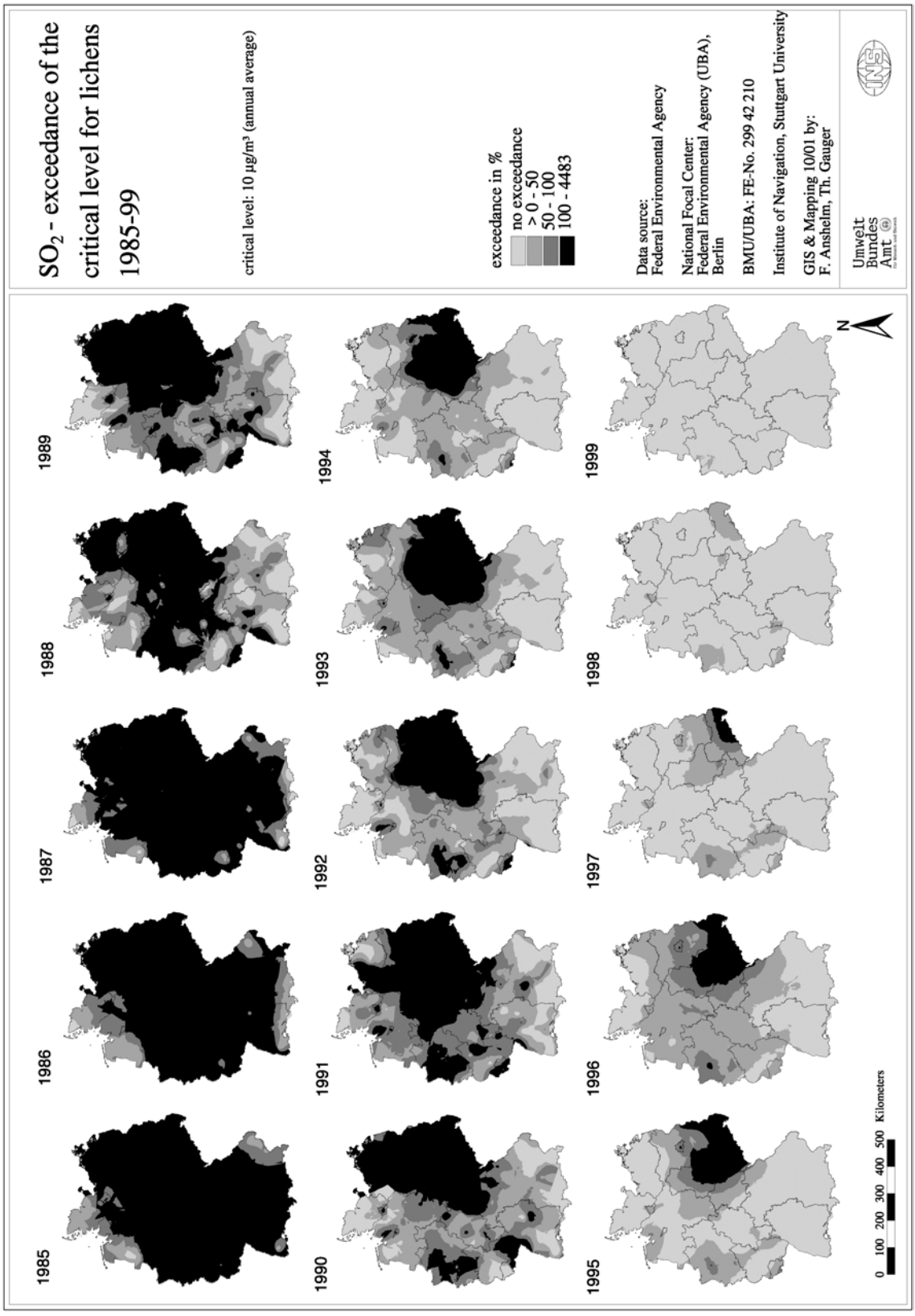
[c] ***A***ccumulated Exposure ***O***ver a ***T***hreshold of ***40*** ppb

[d] daylight hours, e.g. global radiation > 50 W/m²

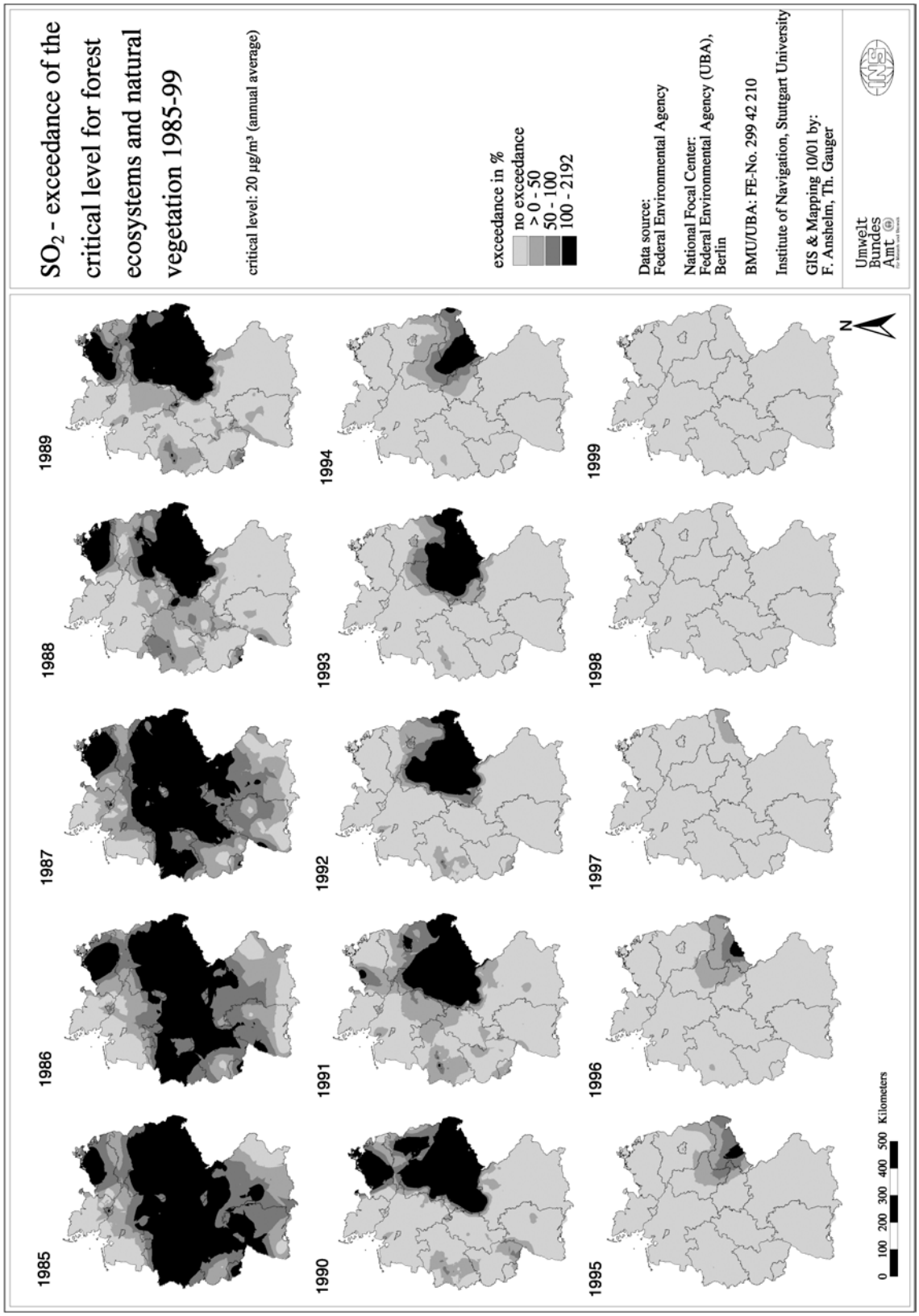
the newly-formed German States too, but in Saxony the exceedances still remained above 100 % till 1997. In 1999 there have been no more exceedances in the newly-formed German States. In North Rhine-Westphalia in 1999 there still have been some small areas where the Critical Level for lichens has been exceeded.

The Critical Level for forest ecosystems and natural vegetation has been exceeded up to 2192 % in the late 1980ies (map 11). The exceedances declined most intensive. In 1998 and 1999 the Critical Level for forest ecosystems and natural vegetation has not been exceeded throughout Germany. In Saxony where SO_2 concentration has been highest in the last decade for the last three years (1998-2000) neither the annual average nor the average of the winter half year has exceeded the Critical Level of 20 µg/m³ (SÄCHSISCHES LANDESAMT FÜR UMWELT UND GEOLOGIE 2000c).

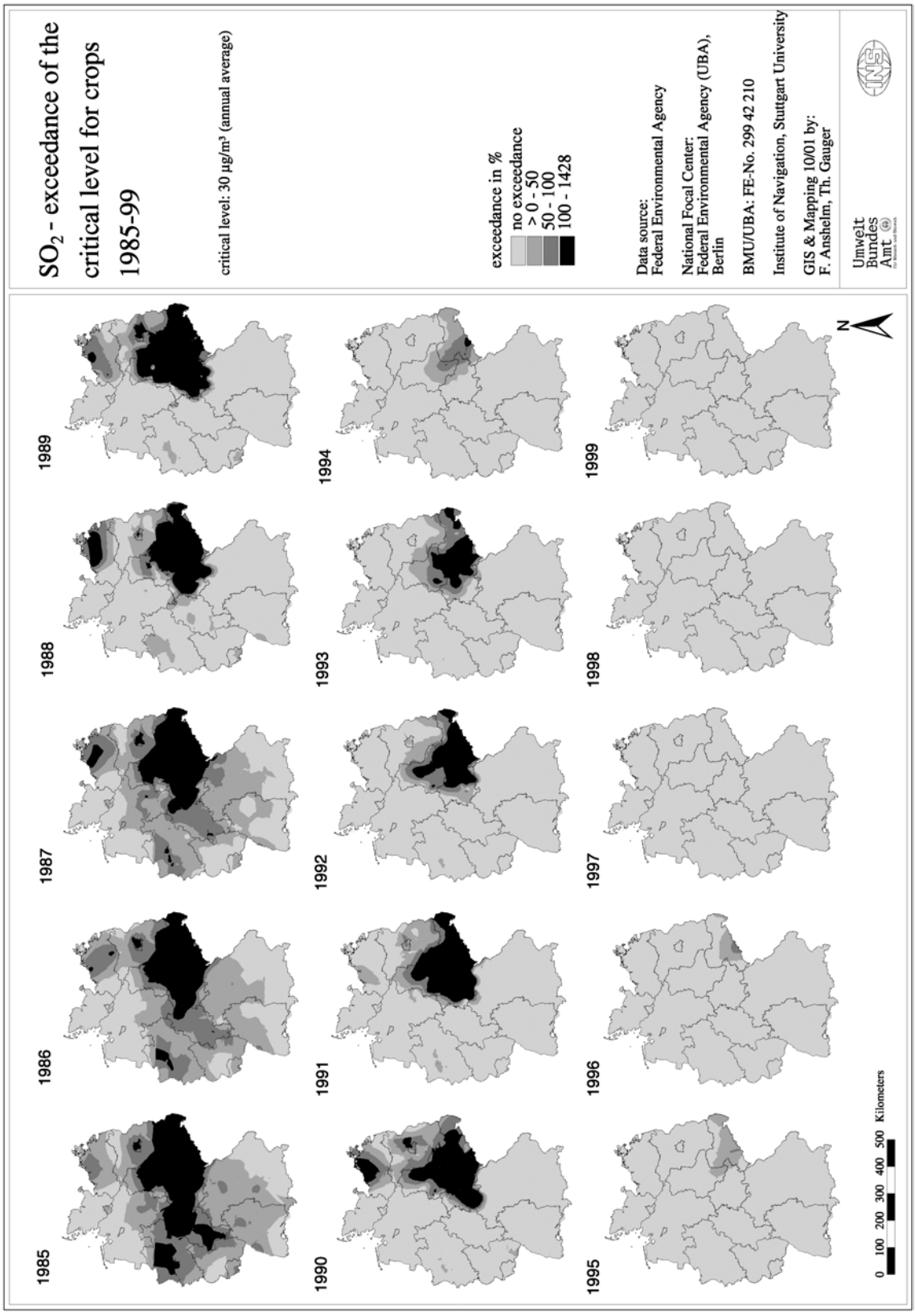
Crops are less sensitive to high concentrations of SO_2. The SO_2 Critical Level for crops therefore is higher than the SO_2 Critical Levels for lichens or forests and natural vegetation. Already in 1985 there have been some regions in Germany where the Critical Level for crops has not been exceeded (among other things Schleswig-Holstein, northern parts of Lower Saxony, south-eastern parts of Bavaria) (map 12). On the other hand there have been regions with exceedances up to 1428 %. Since 1997 there are no exceedances of the Critical Level for crops throughout Germany.



Map: 10: SO_2 – exceedance of the Critical Level for lichens 1985-99.



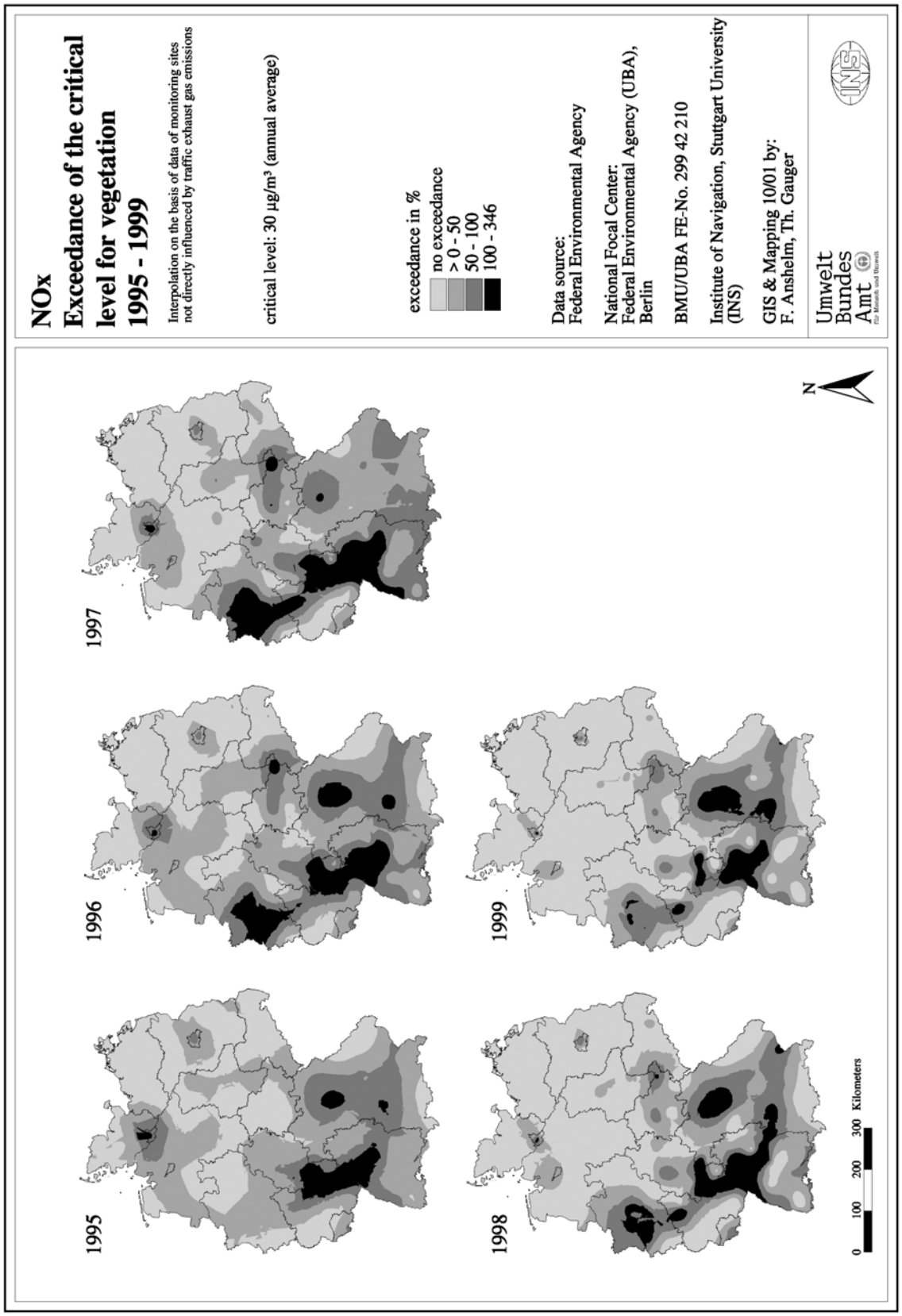
Map 11: SO_2 – exceedance of the Critical Level for forest ecosystems and natural vegetation 1985-99.



Map 12: SO_2 – exceedance of the Critical Level for crops 1985-99.

4.2 Exceedance of the Critical Levels for NO_x

The Critical Levels for NO_x, as established in Egham in 1992 (UN ECE 1992), are still in force. Only the exceedance of the long-term Critical Level for vegetation (annual average of 30 µg/m³) has been mapped until now. The pool of NO_x concentration data is bad. Therefore mapping has been possible for the years 1995-1999 only (map 13). Exceedances reached 346 % in single years. There is no trend detectable. Highest exceedances can be stated in some bigger cities (e.g. Hamburg) or along the Rhine (e.g. Rhine-Main area, Ruhr Basin). But there are also regions where the Critical Level never has been exceeded between 1995 and 1999, for instance wide areas of the eastern and northern German States.



Map 13: NO_X – exceedance of the Critical Level for vegetation 1995-99.

4.3 Exceedance of the Critical Levels for O_3

Since 1992 the Critical Levels for ozone are based on the AOT40 exposure index (Accumulated exposure Over a Threshold of 40 ppb). AOT40 is calculated as the sum of the differences between the hourly concentration of ozone over 40 ppb and 40 ppb (figure 22). Whereas formerly the ozone concentration data of the daylight hours (global radiation > 50 W/m²) or the ozone concentration data between 6 a.m. and 6 p.m. has been used to calculate AOT40 now the ozone concentration data between 8 a.m. and 8 p.m. have to be used. This is an adaptation to the regulations of the 3rd Daughter Directive to the Air Quality Framework Directive (92/62/EC).

The calculation of AOT40 is based on the 1-hour average ozone concentration data. Analogical to mapping ozone concentration only the monitoring data of sites not directly influenced by traffic exhaust gas emissions have been used. Table 4 illustrates the number of datasets suitable for mapping AOT40. AOT40 and the exceedance of the Critical Levels for ozone can be mapped for the years 1990–1999.

Figure 22 illustrates the method of calculating AOT40. On 8 June 1998 at Fichtelberg monitoring site the ozone AOT40 has been 302 ppb.h when using the new reference period (8 a.m. – 8 p.m.). A comparison of the AOT40crops and AOT40forests values calculated with the old (6 a.m. – 6 p.m.) and new (8 a.m. – 8 p.m.) reference period showed that the AOT40 values calculated within the new reference period are somewhat higher than the AOT40 values calculated within the old reference period (AOT40crops ca. 17 % higher and AOT40forests ca. 16 % higher).

Critical Levels for crops and forests are calculated with different time scales. For crops the sum of the daily calculated AOT40 between May and July should not exceed the Critical Level of 3000 ppb.h. The Critical Level for forest ecosystems (AOT40: 10000 ppb.h) should not be exceeded during the six month period April to September.

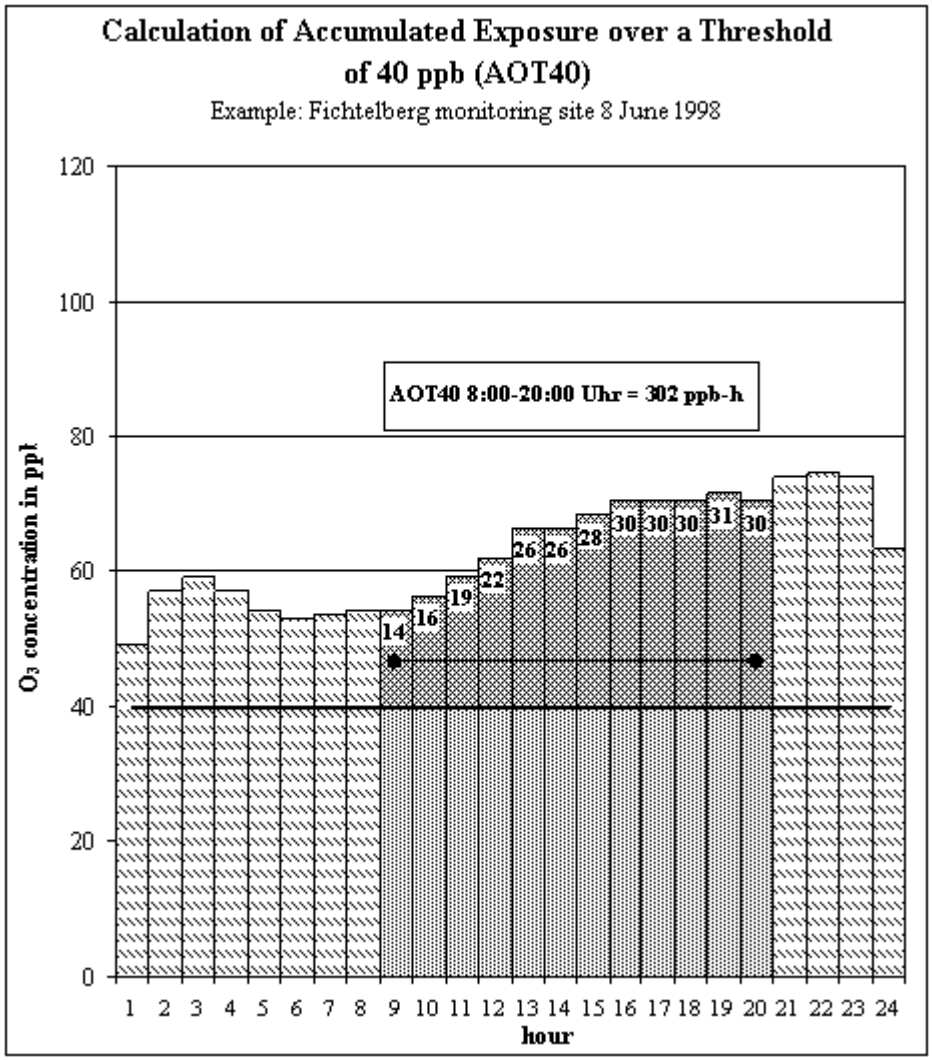
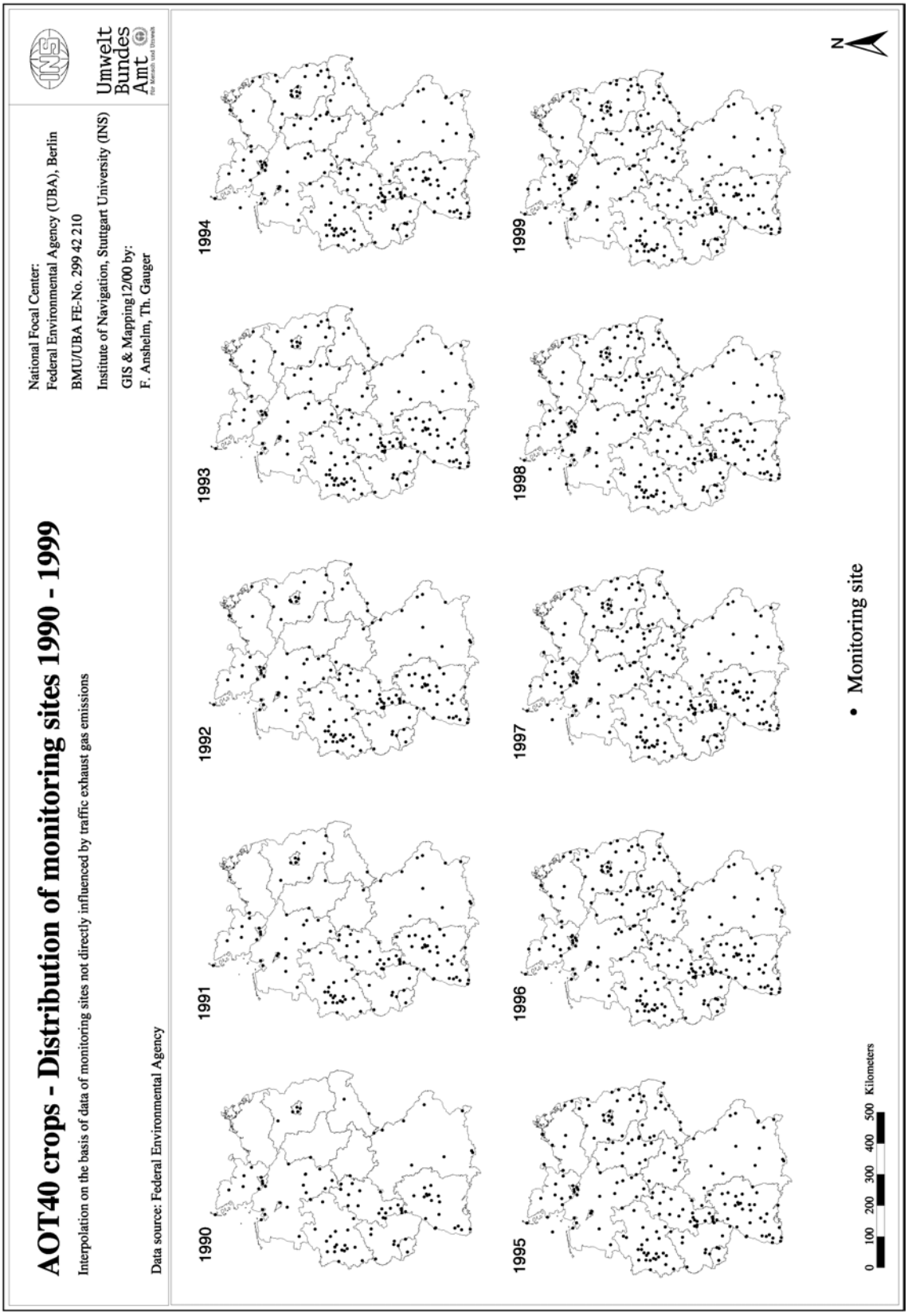
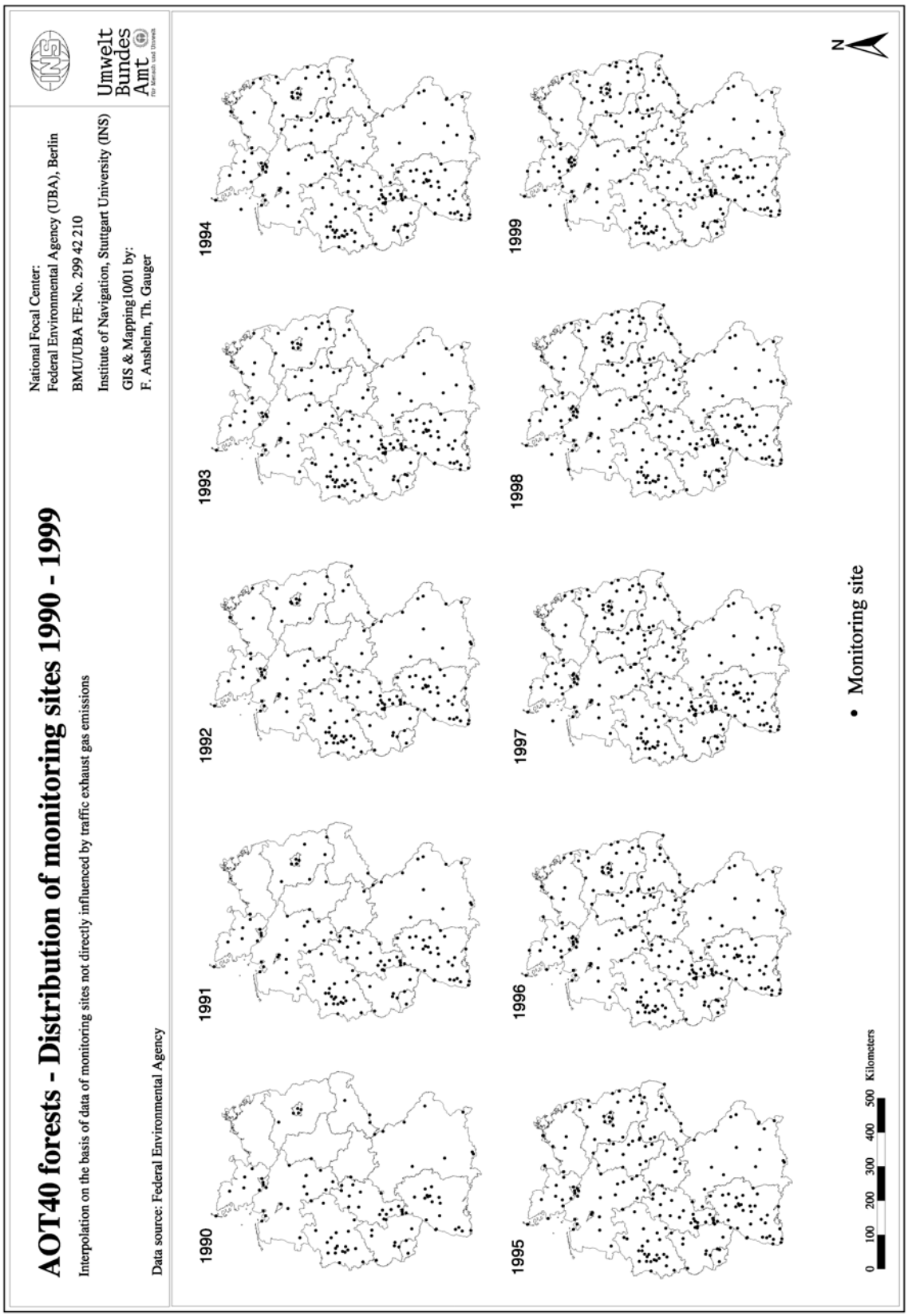


Figure 22: Calculation of AOT40.

The series of ozone monitoring data suitable for calculating and mapping AOT40 and Critical Levels exceedances starts in 1990. In 1990 there are about 110 monitoring sites, which are not directly influenced by traffic exhaust gas emissions (table 4). Till the end of the 1990ies the number of monitoring sites suitable for mapping AOT40crops and AOT40forests increased up to more than 220. Map 14 and 15 illustrate the distribution of the monitoring sites suitable for mapping AOT40crops and AOT40forests. In the early 1990ies there have been some bigger gaps in the monitoring network, especially in the newly-formed German States, in Lower Saxony and in Bavaria. At the latest since 1995 the distribution of monitoring sites is fairly well within Germany.

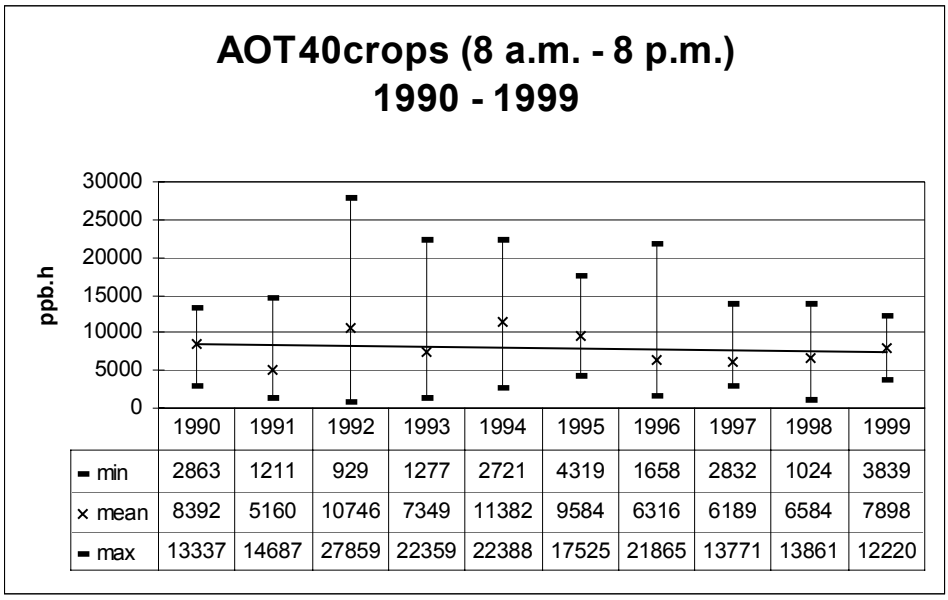


Map 14: AOT40crops – distribution of monitoring sites 1990-99.



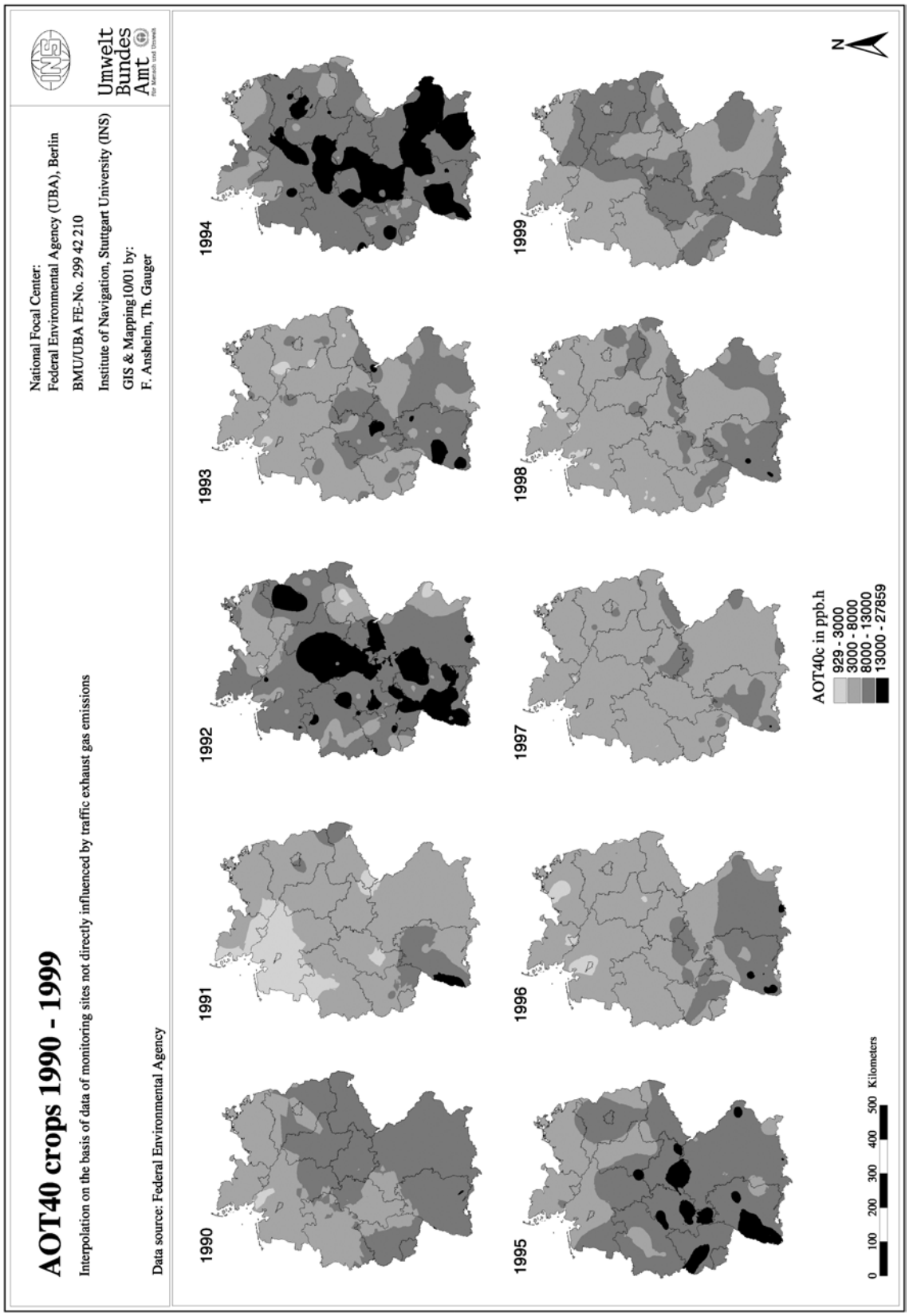
Map 15: AOT40forests – distribution of monitoring sites 1990-99.

AOT40 values have been interpolated using Kriging technique. The quality of the interpolated data strongly depends on the quality of the underlying monitoring network. A high density of monitored data favours good interpolation results. Map 16 illustrates the mapping results for ozone AOT40crops in Germany from 1990 to 1999. AOT40crops varies between 5160 ppb.h (1991) and 11382 ppb.h (1994) (figure 23). The annual maximum values vary between 12220 ppb.h (1999) and 27859 ppb.h (1992). There is no clear trend in AOT40crops data neither in the annual averages nor in the annual maxima. These statements are due to statistical analysis of the AOT40 mapping results and not to statistical analysis of the monitored ozone concentration data itself. Due to the weather conditions in the single years there are considerable fluctuations in AOT40 from year to year. Map 17 shows the 10 years average AOT40crops for the years 1990-1999. Highest values can be stated in the south-western parts of Germany. In the northern parts of Germany AOT40crops values are lower. This is due to differences in climate conditions between the northern and southern parts of Germany. Highest AOT40crops values have been calculated in the southern parts of the Black Forest. This is because of the climate conditions and because of the influence of data from monitoring sites which are located in high altitudes.

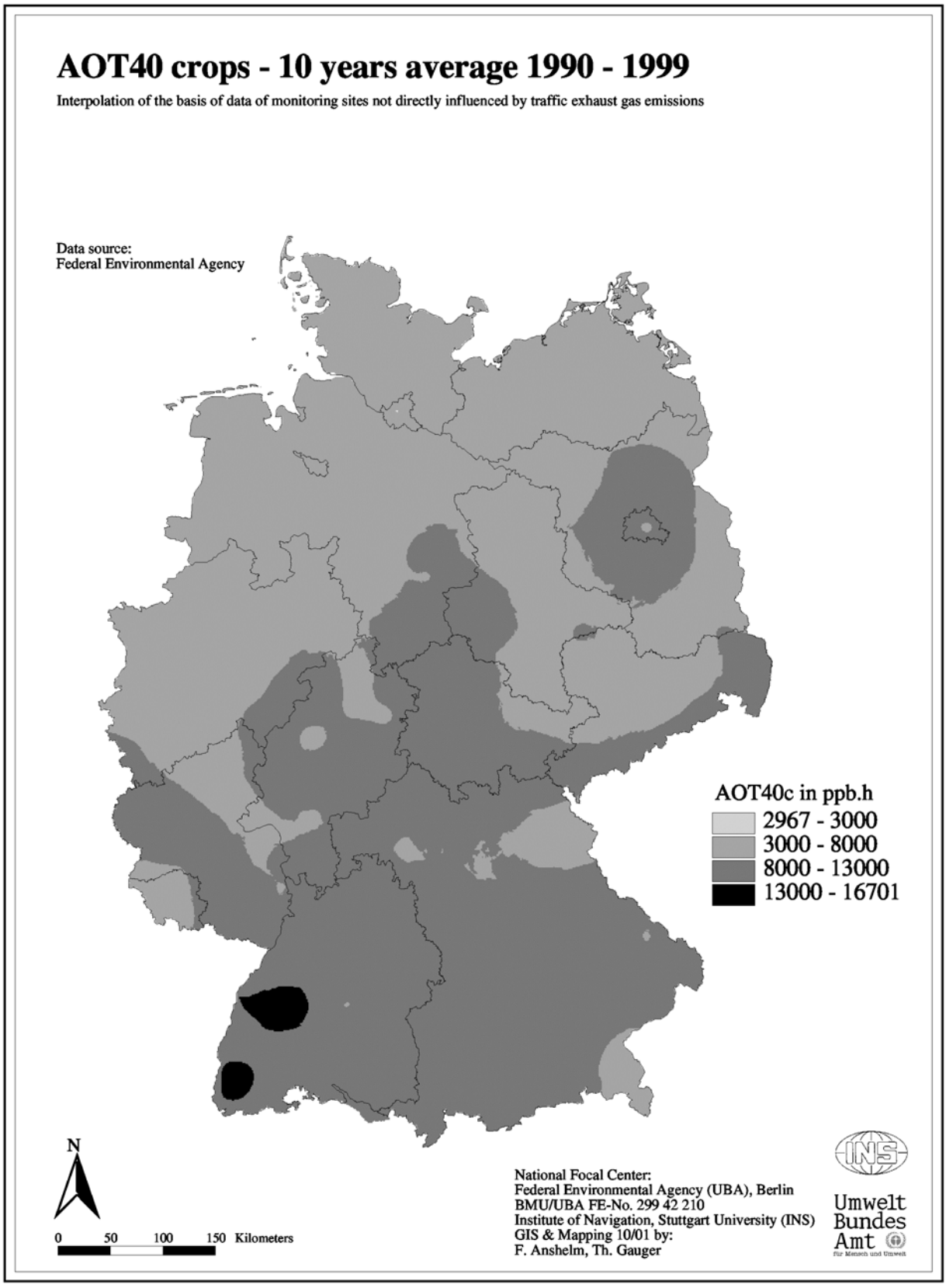


	1990	1991	1992	1993	1994	1995	1996	1997	1998	1999
min	2863	1211	929	1277	2721	4319	1658	2832	1024	3839
mean	8392	5160	10746	7349	11382	9584	6316	6189	6584	7898
max	13337	14687	27859	22359	22388	17525	21865	13771	13861	12220

Figure 23: AOT40crops 1990–1999.

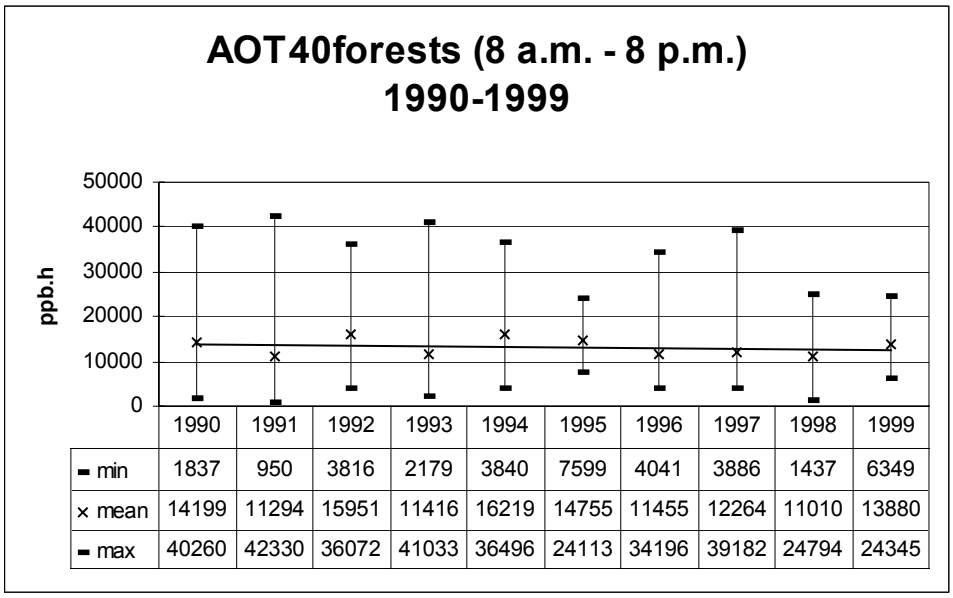


Map 16: AOT40crops 1990-99.


AOT40 crops - 10 years average 1990 - 1999
Interpolation of the basis of data of monitoring sites not directly influenced by traffic exhaust gas emissions
Data source:
Federal Environmental Agency
AOT40c in ppb.h
2967 - 3000
3000 - 8000
8000 - 13000
13000 - 16701
N
0 50 100 150 Kilometers
National Focal Center:
Federal Environmental Agency (UBA), Berlin
BMU/UBA FE-No. 299 42 210
Institute of Navigation, Stuttgart University (INS)
GIS & Mapping 10/01 by:
F. Anshelm, Th. Gauger
INS
Umwelt
Bundes
Amt
für Mensch und Umwelt

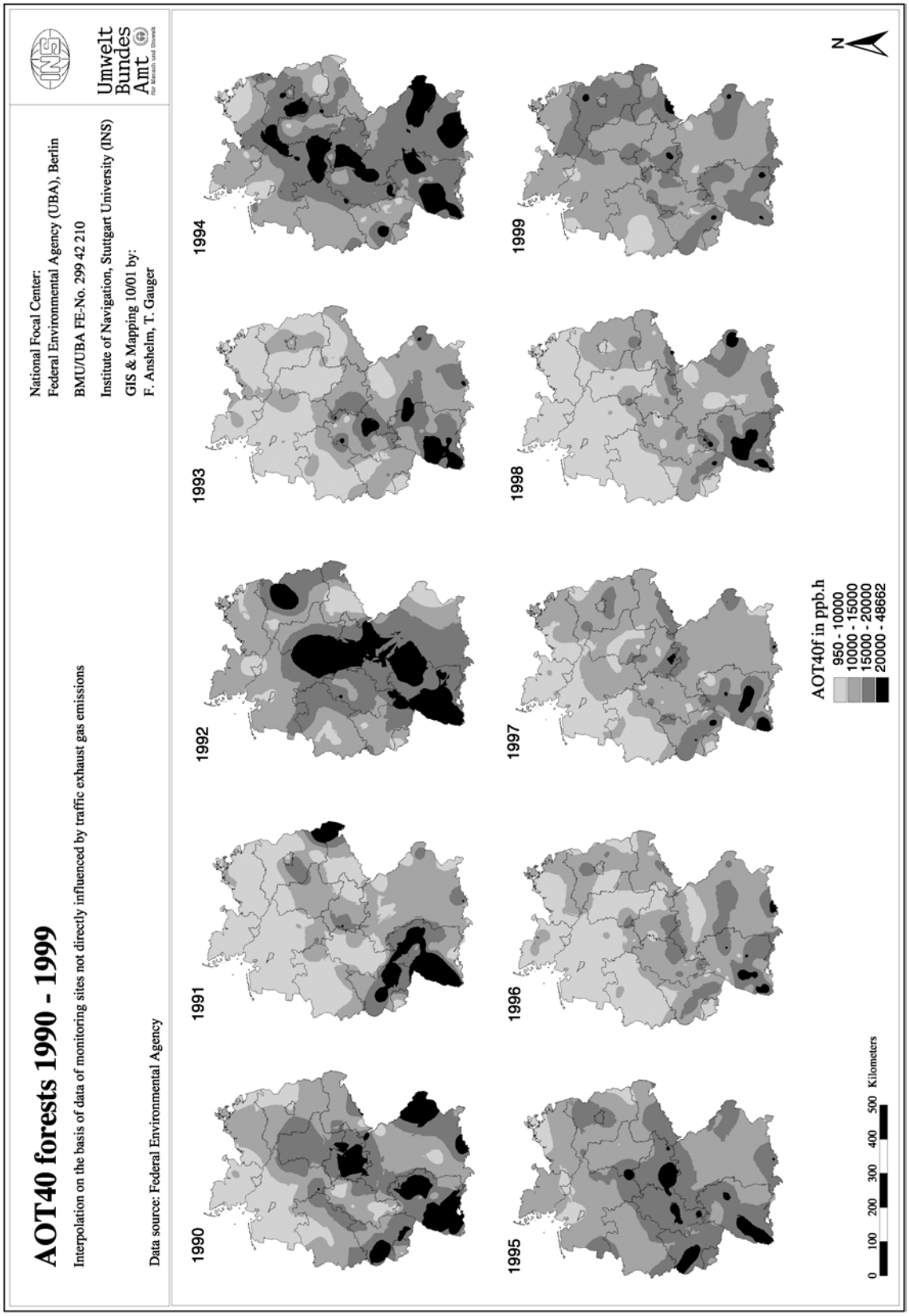
Map 17: AOT40crops – 10-years average 1990-99.

Map 18 illustrates the mapping results for AOT40forests in Germany from 1990 to 1999. Annual average AOT40forests values vary between 11010 ppb.h (1998) and 15951 ppb.h (1992) (figure 24). The annual maximum values vary between 24113 ppb.h (1995) and 42330 ppb.h (1991). There is no clear trend in AOT40forests data neither in the annual averages nor in the annual maxima. These statements are due to statistical analysis of the AOT40 mapping results and not to statistical analysis of the monitored ozone concentration data itself. Due to the weather conditions in the single years there are considerable fluctuations in AOT40 from year to year. Map 19 shows the 10 years average AOT40crops for the years 1990-1999. Highest values can be stated in the southern parts of Germany. In the northern parts of Germany AOT40crops values are lower. This is due to differences in climate between the northern and southern parts of Germany. In the Black Forest AOT40forests is highest, mainly because of the influence of data from monitoring sites which are located in high altitudes. In some regions in the North of Germany the Critical Level of 10000 ppb.h is not exceeded (Bremen, Hamburg, northern Lower Saxony and some parts of North Rhine-Westphalia, Schleswig-Holstein and Mecklenburg-Western Pomerania).

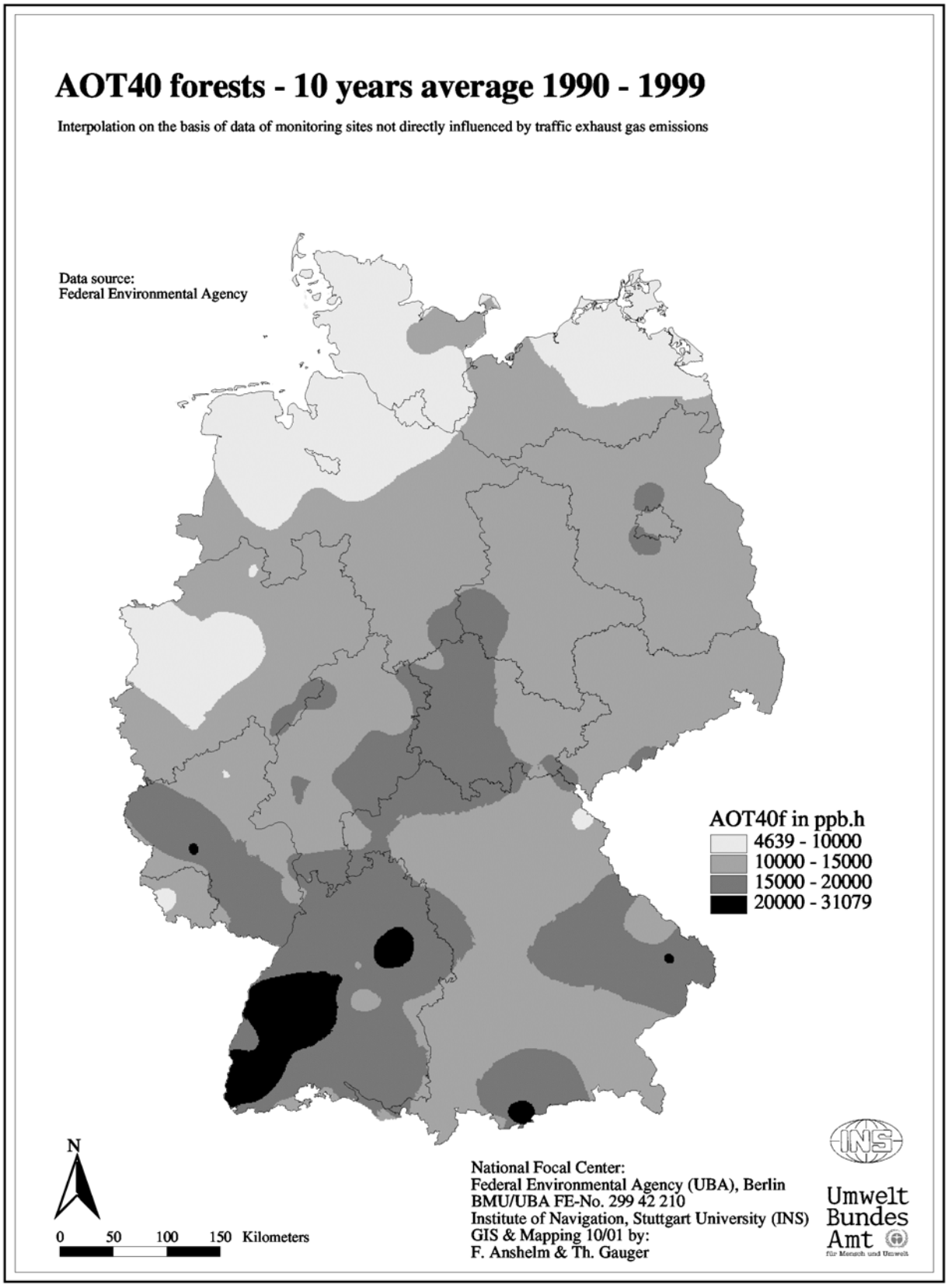


	1990	1991	1992	1993	1994	1995	1996	1997	1998	1999
- min	1837	950	3816	2179	3840	7599	4041	3886	1437	6349
× mean	14199	11294	15951	11416	16219	14755	11455	12264	11010	13880
- max	40260	42330	36072	41033	36496	24113	34196	39182	24794	24345

Figure 24: AOT40forests 1990–1999.



Map 18: AOT40forests 1990-99.


AOT40 forests - 10 years average 1990 - 1999
Interpolation on the basis of data of monitoring sites not directly influenced by traffic exhaust gas emissions
Data source:
Federal Environmental Agency
AOT40f in ppb.h
4639 - 10000
10000 - 15000
15000 - 20000
20000 - 31079
N
0 50 100 150 Kilometers
National Focal Center:
Federal Environmental Agency (UBA), Berlin
BMU/UBA FE-No. 299 42 210
Institute of Navigation, Stuttgart University (INS)
GIS & Mapping 10/01 by:
F. Anshelm & Th. Gauger
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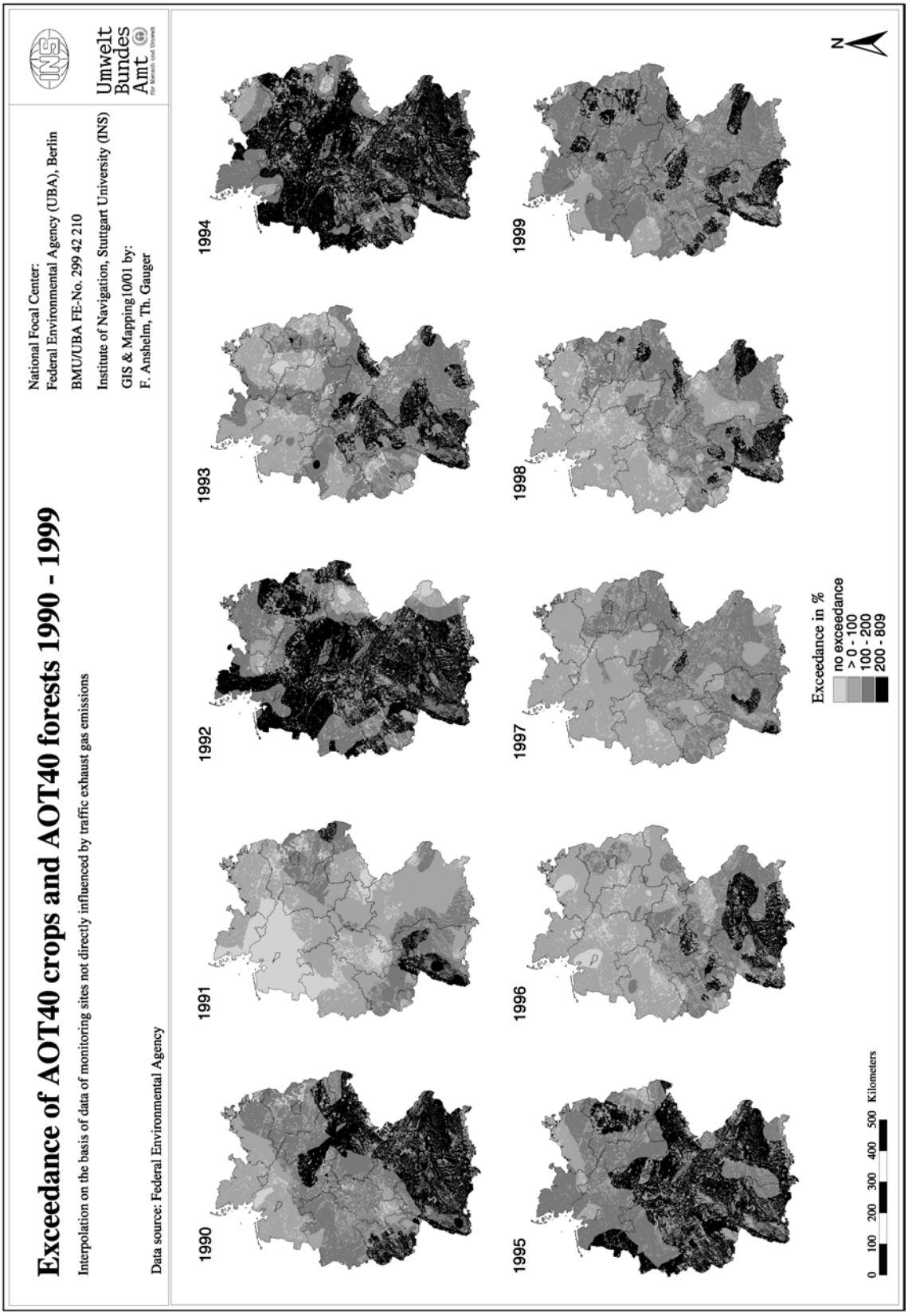
Map 19: AOT40forests – 10-years average 1990-99.

Map 20 illustrates the percental exceedance of AOT40crops and AOT40forests for the years 1990-1999. There are exceedances up to 809 % detectable, but in every year there are some places where the Critical Levels are not exceeded. Map 21 shows the 10 years average exceedance of Critical Levels 1990-1999. There are exceedances up to 428 % and only very small areas with no exceedance. Highest exceedances can be stated in the southern and central parts of Germany.

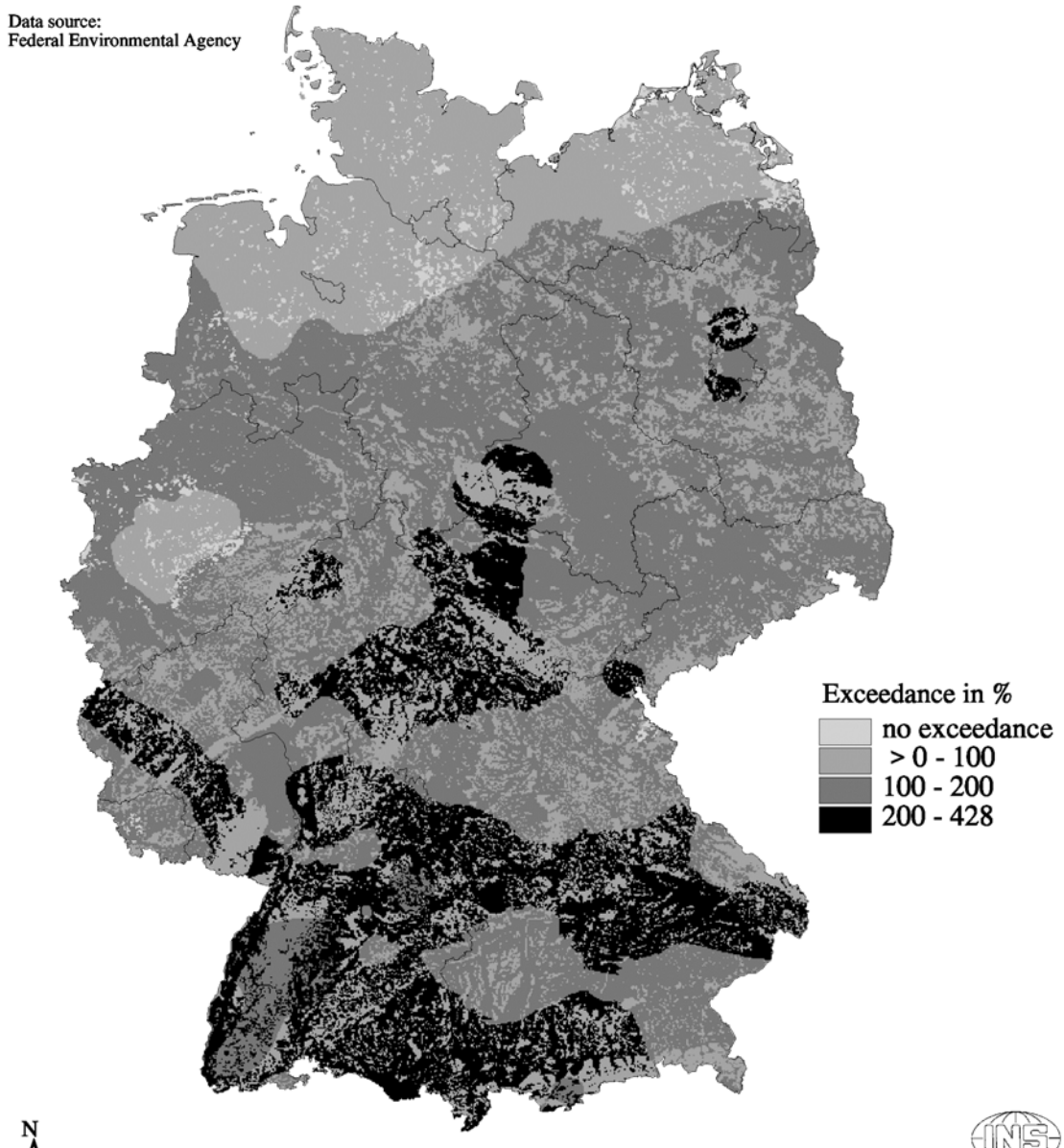


Map 20: Exceedance of AOT40crops and AOT40forests 1990-99.

Exceedance of AOT40 crops and AOT40 forests
10 years average 1990 - 1999

Interpolation of the basis of data of monitoring sites not directly influenced by traffic exhaust gas emissions

Data source:
Federal Environmental Agency



National Focal Center:
Federal Environmental Agency (UBA), Berlin
BMU/UBA FE-No. 299 42 210
Institute of Navigation, Stuttgart University (INS)
GIS & Mapping 10/01 by:
F. Anshelm, Th. Gauger

Umwelt Bundes Amt
für Mensch und Umwelt

Map 21: Exceedance of AOT40crops and AOT40forests – 10-years average 1990-99.

5 Mapping ozone AOT60 and WHO Guideline Level for the protection of human health

"A Critical Level of ozone for human health is represented by the WHO Air Quality Guideline level for ozone of 120 µg/m³ (moving 8-hours average). Within UN ECE context and in close collaboration with the World Health Organization's Regional Office for Europe (WHO/EURO), a Critical Level expressed as an AOT60 (accumulated exposure over a threshold of 60 ppb), i.e. 120 µg/m³, calculated over one year, was adopted as a surrogate for the WHO Air Quality Guideline level for the purpose of integrated assessment modelling" (UN ECE Gothenburg Protocol). Both standards – WHO Air Quality Guideline level for ozone and ozone AOT60 have been calculated and mapped for the years 1990 to 1999, in order to identify areas at risk, where the standards are exceeded.

5.1.1 Health effects of ground-level ozone

High levels of ozone damage the lungs and are particularly dangerous to vulnerable groups such as children, asthmatics, others with breathing disorders, and the elderly. High ozone concentrations mean that these groups should avoid exposure and exercise. As the pollution levels of ozone increase so exercise by all becomes more dangerous. Recent findings from the USA suggest that the long-term exposure of lower levels of ozone also inflict a lasting damage on the lungs (figure 25).

Ozone toxicity occurs in a continuum in which higher concentrations, longer exposure duration, and greater activity levels during exposure cause greater effects. When inhaled, even at very low levels, ozone can

- cause acute respiratory problems
- aggravate asthma
- cause significant temporary decreases in lung capacity of 15 to over 20 percent in some healthy adults
- cause inflammation of lung tissue
- impair the body's immune system defences, making people more susceptible to respiratory illnesses, including bronchitis and pneumonia

There is, however, a great variability in individual responsiveness that is not yet understood. Considering the severity of health effects of elevated ground-level ozone concentrations several international organisations deal with this problems.

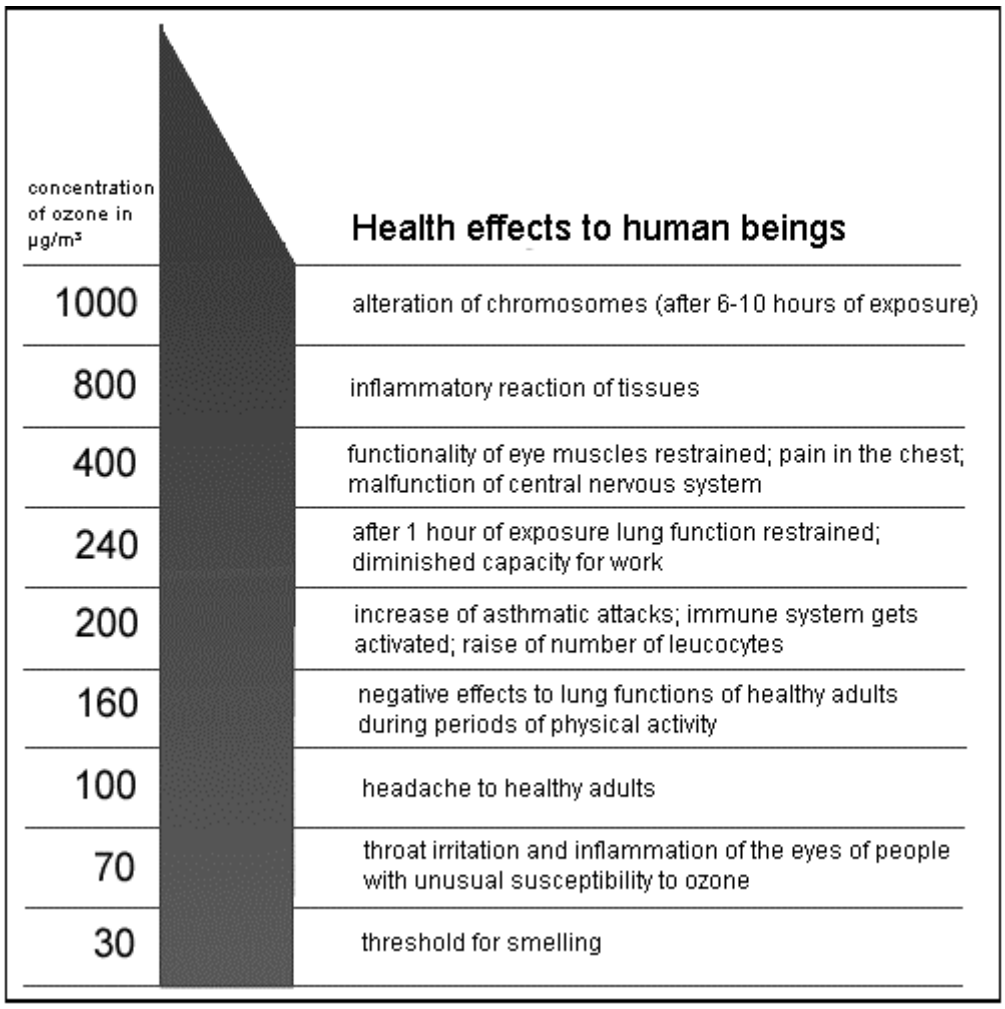


Figure 25: Ozone health effects to human beings.

5.1.2 Ozone and health effects – guidelines

Already in 1958, **WHO (World Health Organisation)** recognised, that air pollution is a threat to the health and well-being of peoples throughout the world. As a consequence, WHO has taken its first steps to marshal the facts and to suggest procedures by which preventive and remedial action may be taken by its member countries, before serious harm is done to the health of their people. Guidelines were defined as sets of concentrations and exposure times that are associated with specific effects of varying degrees of air pollution on man, animals, vegetation

and on the environment in general. For many of the „classic“ pollutants (SO_2, SPM[5], CO and photochemical oxidants) guidances were formulated which based on controlled exposure studies, or on epidemiological studies which demonstrated a threshold of effect. These attempts culminated in 1987 in the publication of the Air Quality Guidelines for Europe for a much extended set of air pollutants.

During the development of the 1987 WHO Guidelines, emphasis was placed on specifying the guidelines in terms of a concentration and averaging time, which would define an exposure unlikely to produce adverse effects, even in the majority of those members of groups with increased sensitivity to the pollutant in question. The guidelines were statements of levels of exposure at which, or below which, no adverse effects can be expected.

In the revised version of the WHO Air Quality Guidelines for Europe (WHO 1999), a similar approach was applied as in the 1987 Air Quality Guidelines. The principles applied were those developed by the Working Group on Effects under the Convention on Long-Range Transboundary Air Pollution of the UN ECE, and the evaluations were carried out jointly with that group. Critical Levels and Critical Loads were derived. A Critical Level of ozone for human health is represented by the WHO Air Quality Guideline level for ozone (moving 8-hours average of 120 µg/m³).

Under the **UN ECE Convention on Long-Range Transboundary Air Pollution** the Working Group on Effects developed principles to evaluate air pollution effects on the environment, including human health. **Critical Levels** are defined as concentrations of pollutants in the atmosphere above which direct adverse effects on receptors, such as human beings, plants, ecosystems or materials, may occur according to present knowledge. **Critical Load** is a quantitative estimate of an exposure to one or more pollutants below which significant harmful effects on specified sensitive elements of the environment do not occur according to present knowledge. Within the „Protocol to the 1979 Convention on Long-Range Transboundary Air Pollution to abate acidification, eutrophication and ground-level ozone“ **(Gothenburg Protocol)** a Critical Level expressed as an **AOT60** (accumulated exposure over a threshold of 60 ppb), i.e. 120 µg/m³, calculated over one year, was adopted as a surrogate for the WHO Air Quality Guideline level for the purpose of integrated assessment modelling. It is important to stress that this AOT60 surrogate indicator has been introduced purely for practical modelling reasons. Given the current knowledge on health effects it is not possible to link any AOT60 value larger than zero with a

[5] suspended particulate matter

certain risk to human health. The only possible interpretation is that if the AOT60 is above zero, the WHO criterion could be exceeded at least once during the period in question.

Under the **Air Quality Framework Directive (96/62/EC)** a proposal for a new Ozone Directive (3rd Daughter Directive) has been put forward by the Commission of the EU. In this new proposal long-term objectives and target values for the protection of human health and vegetation are defined and based on the revised guidelines of WHO. The target values should be attained, as far as possible, in 2010. For the protection of human health **the maximum daily 8-hours average of 120 µg/m³ should not be exceeded on more than 20 days per calendar year averaged over three years**. The long-term objective for the protection of human health is no exceedance of the daily 8-hour mean of 120 µg/m³ within a calendar year. In addition, the proposed EC 3rd Ozone Directive sets information and general alert thresholds. Whereas for humans an exposure period of 8 hours is considered, for the protection of vegetation the relevant period extends over the whole growing season (three months). AOT40 should not exceed 6000 µg/m³ (long-term-objective). Based on the gap between current ozone levels and the targets, one might say that the targets for protection of vegetation will be more difficult to meet than the human health related targets. The guidelines of the coming 3rd Ozone Directive of the EU will have to be put into national legislation like it has been done with the guidelines of the Ozone Directive 92/72/EWG (= 22.BImSchV), which is still valid.

5.1.3 Calculation and mapping of AOT60 and WHO Air Quality Guideline level for the protection of human health

Figure 26 illustrates the procedure of mapping AOT60 and WHO Air Quality Guideline level for the protection of human health. Calculations are based on air pollutant monitoring data (1-hourly averages) of at least 110 (1990) German monitoring sites. Since 1995 data of more than 200 monitoring sites are available. Table 4 illustrates the number of datasets suitable for mapping AOT60 and WHO Air Quality Guideline level for the protection of human health for the years 1990 to 1999. The number of datasets suitable for mapping AOT60 is different to the number of datasets suitable for mapping WHO Air Quality Guideline level because of different ways to eliminate missing values. The method of calculating AOT60 and WHO Air Quality Guideline level will be described in chapter 5.1.4 and 5.1.5. The data of the monitoring sites located nearby streets are not suitable for mapping purposes, because of their small area of representativeness. The area of representativeness of monitoring sites depends on the surrounding of the site. In areas with high vehicle exhaust emissions concentrations of ozone are very low due

to the reaction of ozone with nitric oxide (NO), which rapidly generates nitrogen dioxide (NO_2). Therefore, the area of representativeness of monitoring sites located nearby streets is rather small. For mapping purposes only the data of monitoring sites not directly affected by traffic emissions have been used. Subsequently, Kriging interpolation techniques are applied to calculate grids for the whole area of Germany out of the point datasets of the monitoring sites. The output resolution of the grids is 1·1 km². Finally, the validity of the mapping results is checked and a statistical evaluation of the mapping results is carried out.

5.1.4 Mapping WHO Air Quality Guideline level

Figure 27 illustrates the calculation of the exceedance of the WHO Air Quality Guideline level. Firstly, 8-hours moving averages are calculated. There are twenty-four 8-hours moving averages for every day. The end of a 8-hours period decides to which day the 8-hours average is assigned to. 8-hours periods which end later than midnight are assigned to the morrow.

Acute effects on public health are likely to be small as long as the maximum moving 8-hours average concentration of 120 µg/m³ (= 60 ppb) is not exceeded. If there is at least one 8-hours average higher than 120 µg/m³ within a day, this day is marked as a day of exceedance of the WHO Air Quality Guideline level. The exceedance of the WHO Air Quality Guideline level is calculated for every day of the year and each monitoring site not directly affected by vehicle exhaust gas emissions within Germany.

The data of monitoring sites located nearby streets have to be removed from the dataset, because of their small area of representativeness. On the basis of the remaining monitoring sites Kriging interpolation techniques have been applied to calculate grids for the whole area of Germany. The output resolution of the grids is 1·1 km².

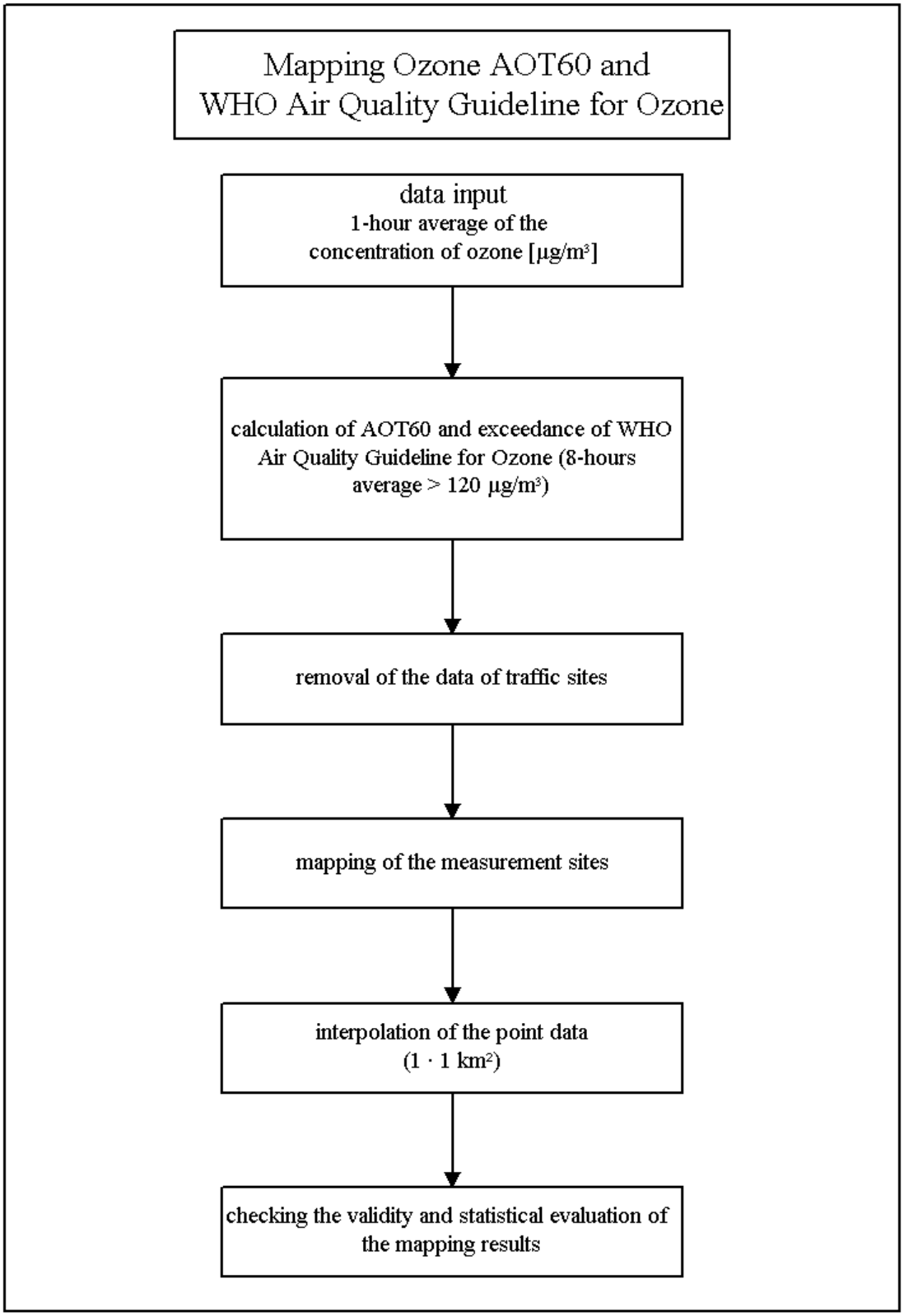


Figure 26: Mapping AOT60 and WHO Air Quality Guideline level for ozone.

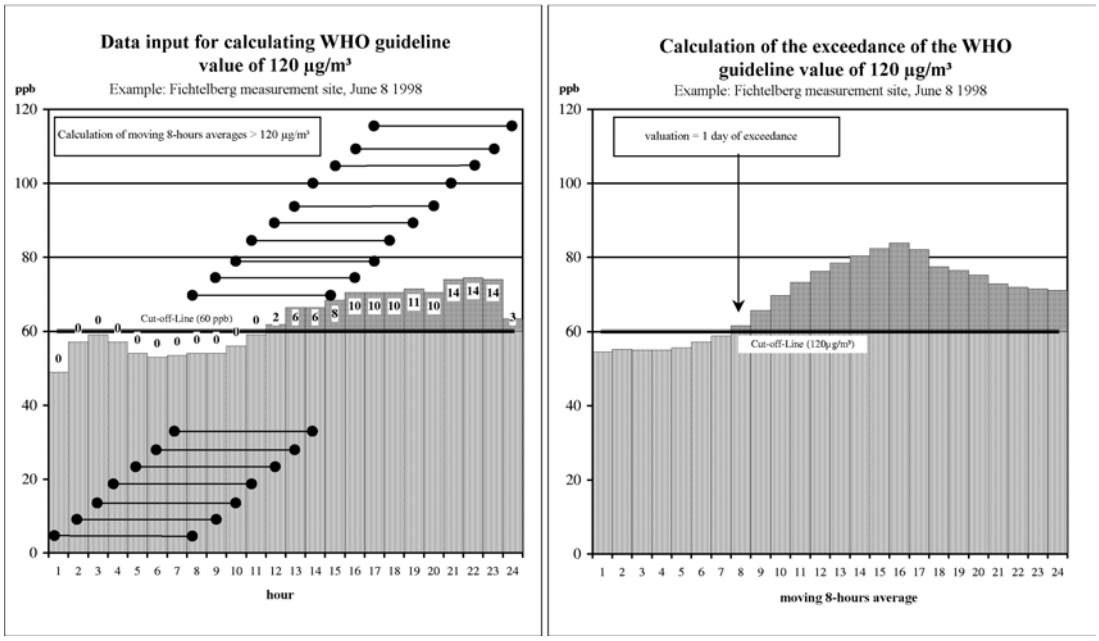
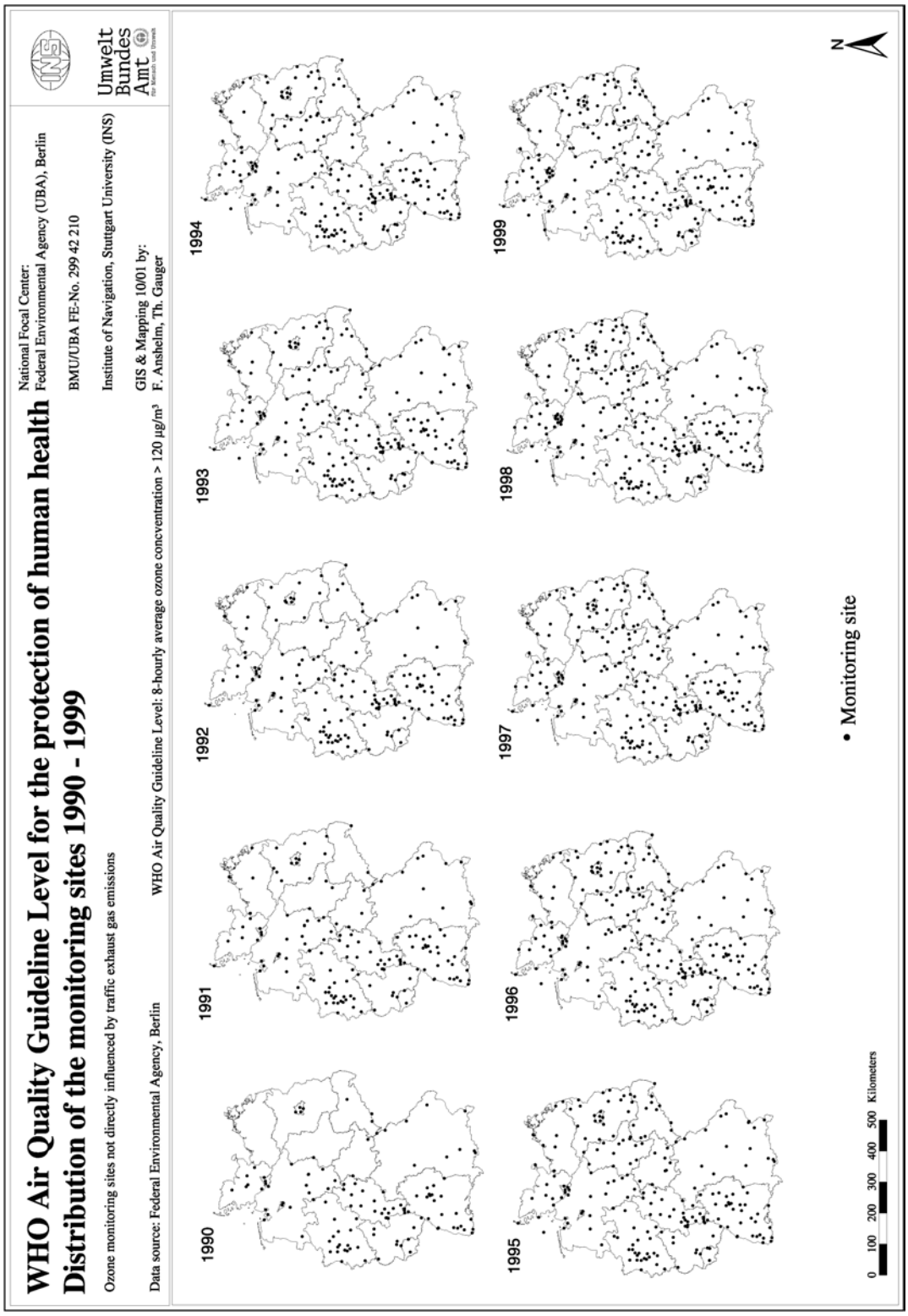
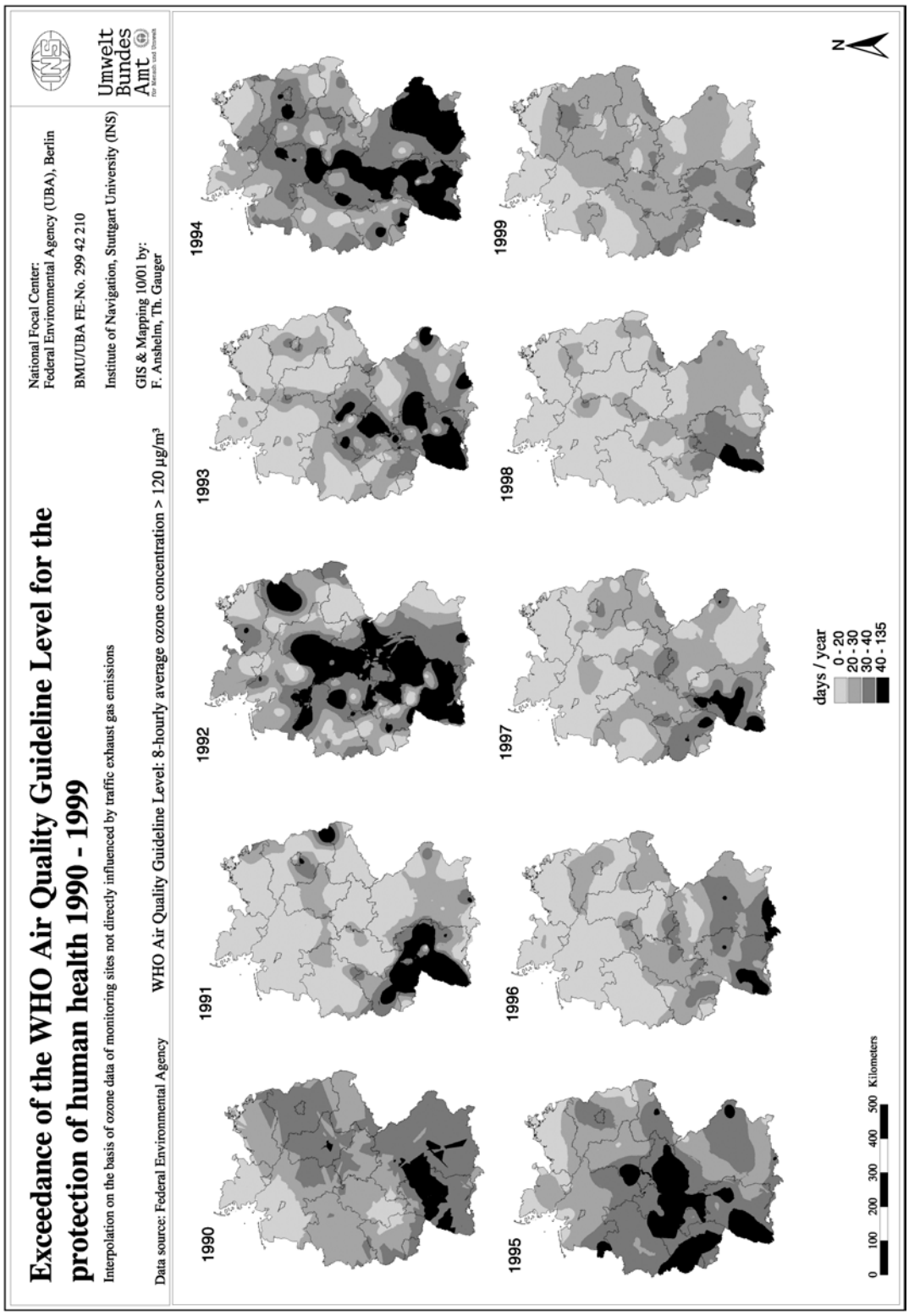


Figure 27: Data input and calculation of the exceedance of the WHO Air Quality Guideline level for the protection of human health.

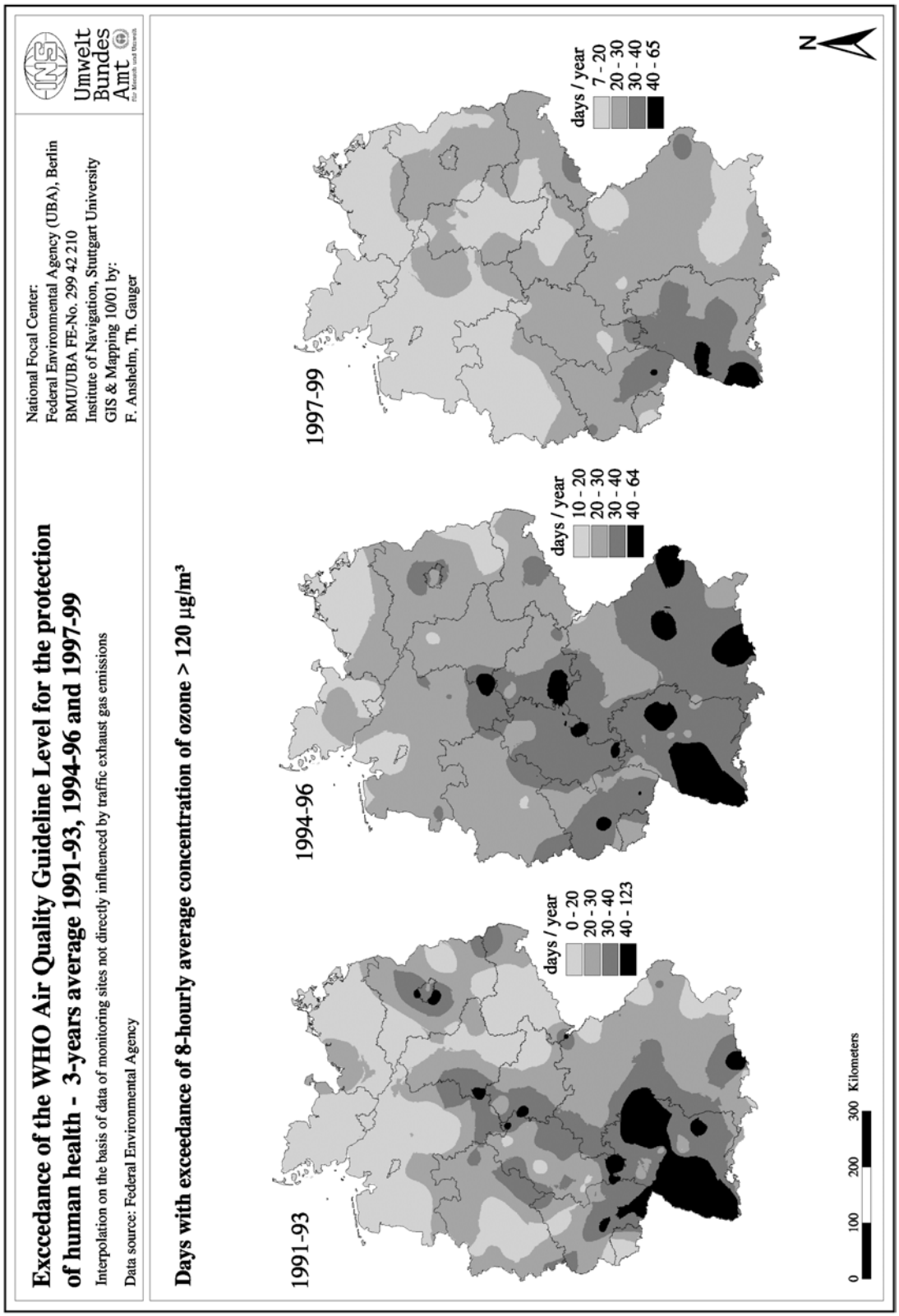
Map 22, 23 and 24 illustrate the mapping results for the years 1990-1999. In south-west Germany and in higher altitudes there are more days with exceedance of the WHO criterion than in the North and in lower altitudes. The highest number of days with exceedance of the WHO criterion are found in 1992. In the late 1990ies the number of days with exceedance is relatively low. Exceedances of more than 80 days are restricted to small areas in higher altitudes (Black Forest, The Alps). For the protection of human health the maximum daily 8-hours average of 120 µg/m³ should not be exceeded on more than 20 days per calendar year averaged over three years. Map 24 illustrates the exceedances for three years averages. There are high exceedances in the south-western parts of Germany and slight exceedances round the northern regions. There are striking differences between the three periods. In the period from 1997 to 1999 there are relatively small exceedances of the 20 days criterion.



Map 22: WHO Air Quality Guideline Level for the protection of human health – distribution of the monitoring sites 1990-99.

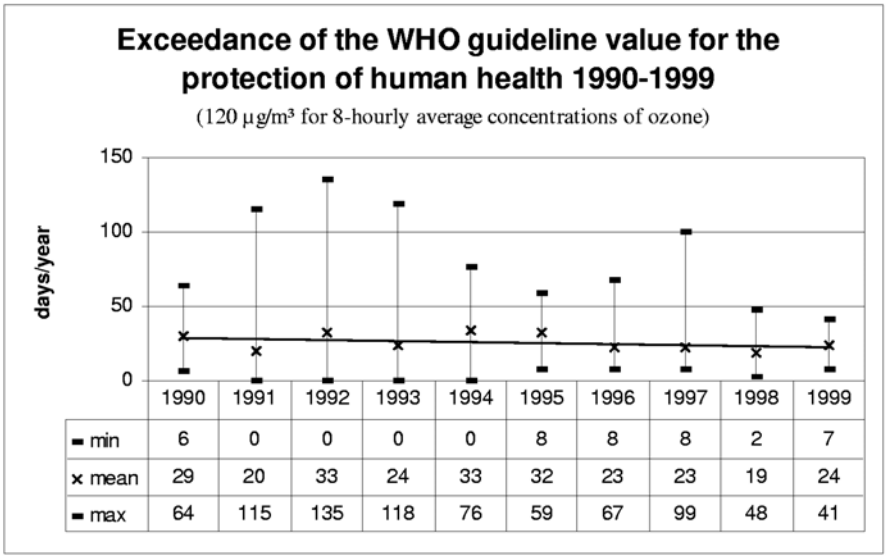


Map 23: Exceedance of the WHO Air Quality Guideline Level for the protection of human health 1990-99.



Map 24: Exceedance of the WHO Air Quality Guideline Level for the protection of human health – 3-years average 1991-93, 1994-96 and 1997-99.

Figure 28 illustrates the results of a statistical evaluation of the mapping results of the years 1990 -1999. There is a decline of the maximum values and a tender downward trend of the annual averages. Further investigations will be necessary to verify whether there is a reliable trend of the annual averages.



	1990	1991	1992	1993	1994	1995	1996	1997	1998	1999
min	6	0	0	0	0	8	8	8	2	7
mean	29	20	33	24	33	32	23	23	19	24
max	64	115	135	118	76	59	67	99	48	41

Figure 28: Exceedance of the WHO Air Quality Guideline level for the protection of human health 1990-1999.

The method of calculating the WHO Air Quality Guideline level for the protection of human health is totally different to the method used to calculate ozone AOT60, whereas the mapping methods are identical. This has to be considered when trying to compare the mapping results.

5.1.5 Mapping ozone AOT60

The method of calculating and mapping AOT60 is almost identical to the method used to calculate and map AOT40. Nevertheless there are some differences. First of all, the cut-off-line for calculating the accumulated exposure is different - 60 ppb instead of 40 ppb. In contrast to AOT40 the accumulated exposure over a threshold of 60 ppb considers the air concentration data of the whole day and the whole year.

Figure 29 illustrates the method of calculating AOT60 for one day and one monitoring site. There are 24 one-hour average concentrations per day (in ppb). The sum of the ppb.hours above the cut-off-line of 60 ppb forms the AOT60 for this particular day. Subsequently, the diurnal AOT60 values of the monitoring sites are summed up to form annual AOT60 values.

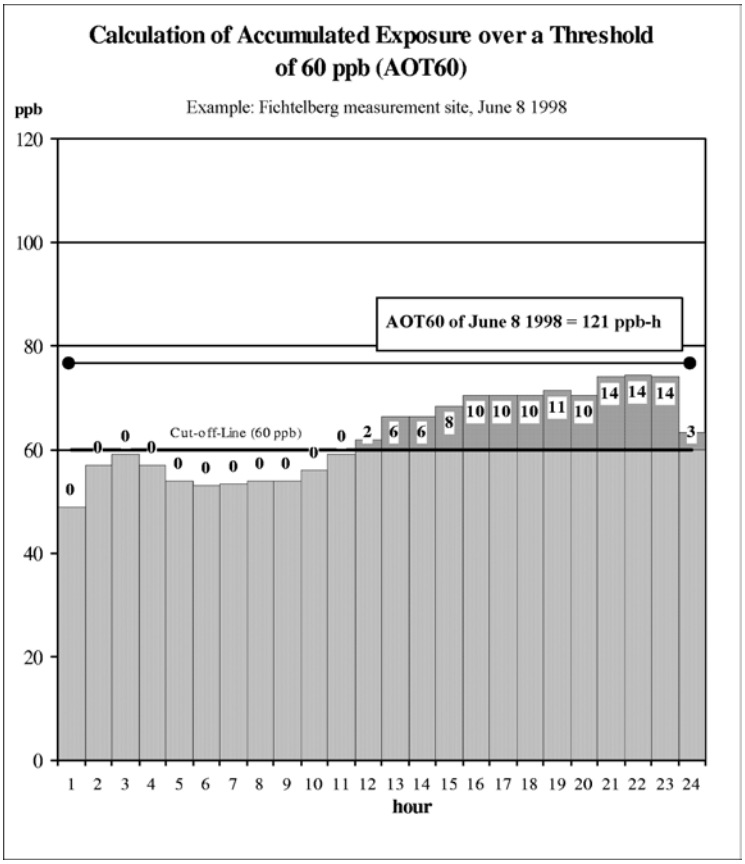
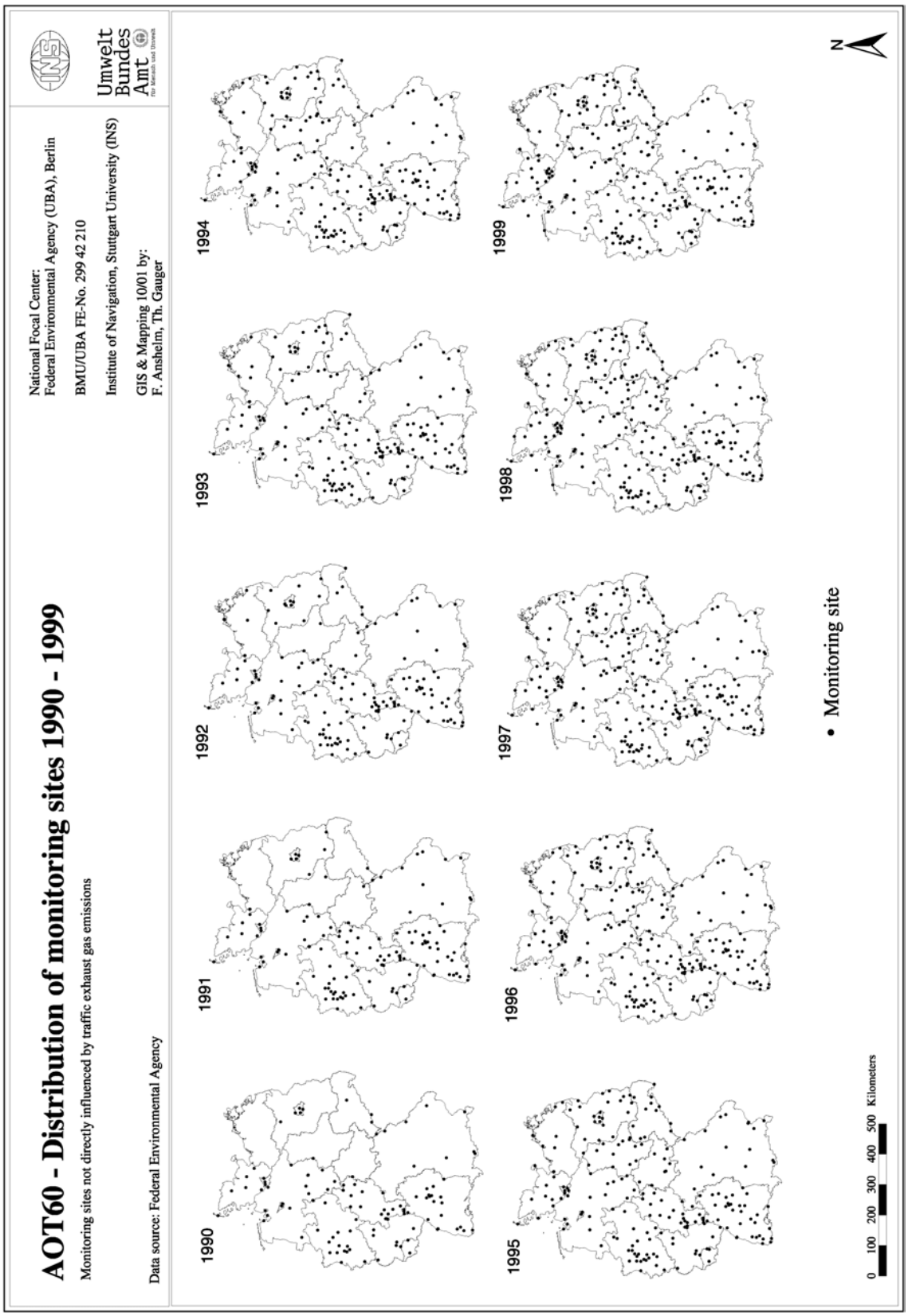
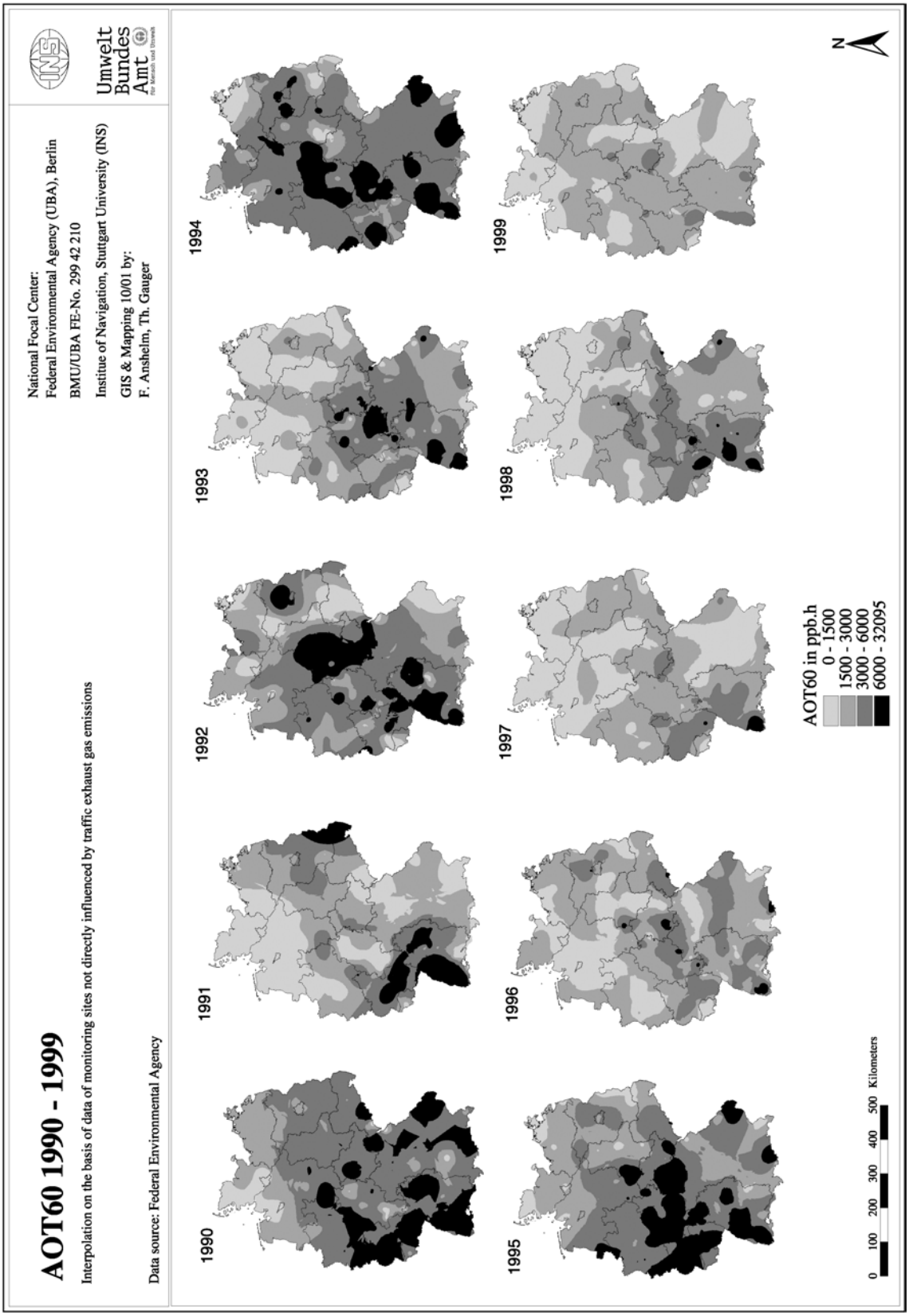


Figure 29: Calculation of Accumulated exposure Over a Threshold of 60 ppb (AOT60).

Map 25 and 26 illustrate the mapping results for the years 1990-1999. Situation changes irregularly from year to year due to weather conditions and concentration of precursor compounds (VOC, NO_x). Sunny, warm weather and high levels of precursor compounds as well as ozone transport into regions remote from NO emission areas favour high concentrations of ozone and consequently high AOT60 values. The frequency of high AOT60 values is higher in the south-western parts of Germany than in the North or in the East. Highest AOT60 values are found at remote mountain. There are some years with high ozone concentration levels, e.g. 1990, 1992, 1994 and 1995. In the late 1990ies the area with very high AOT60 values diminished.

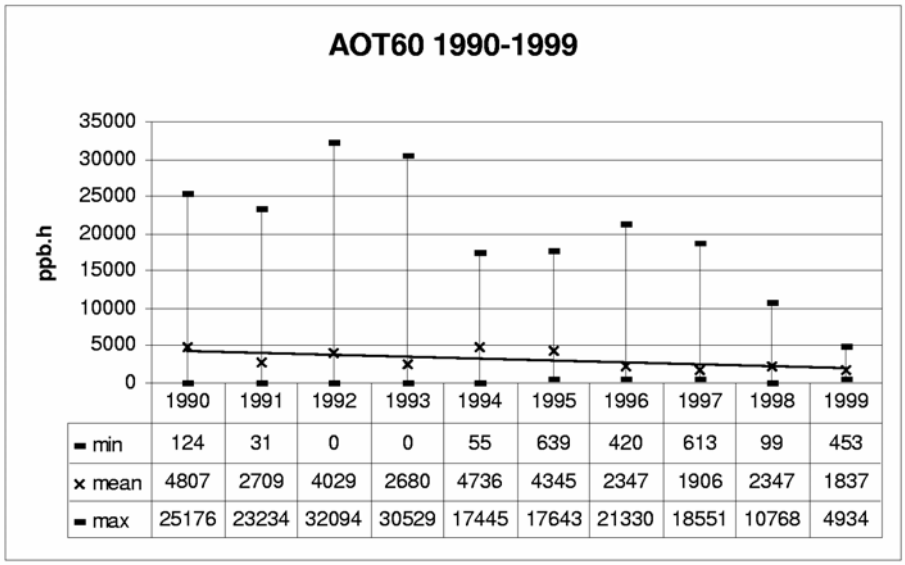


Map 25: AOT60 – distribution of the monitoring sites 1990-99.



Map 26: AOT60 1990-99.

A statistical evaluation of the mapping results has been carried out for the years 1990 -1999 (figure 30). There is a decline of the maximum AOT60 and a tender downward trend referring to the annual average AOT60. There is a need for further investigations to verify, whether there is a reliable trend. Maximum AOT60 values are found at remote mountain areas of high elevation. AOT60 of more than 20000 ppb.h are restricted to these areas (Black Forest, The Alps). The ozone concentration data of these high-elevated monitoring sites are influenced by high ozone concentrations of the ozone reservoir layer (ca. 300 - 1500 m above ground).



	1990	1991	1992	1993	1994	1995	1996	1997	1998	1999
– min	124	31	0	0	55	639	420	613	99	453
× mean	4807	2709	4029	2680	4736	4345	2347	1906	2347	1837
– max	25176	23234	32094	30529	17445	17643	21330	18551	10768	4934

Figure 30: Statistical evaluation of AOT60 mapping results for the years 1990-1999.

6 AOT40 Level II concept

The ozone Critical Levels actually in force have been defined on the basis of experimental data using a Level I approach (AOT40crops and AOT40forests). This Critical Levels consider exposition of receptors to ozone and not ozone fluxes into leaves and needles, despite the latter one correlates better with the effects to plants. The Level I approach is attractive because of its simplicity, but it does not consider any environmental factor that may influence a plant's sensitivity to ozone. The Level I approach therefore is not the most suitable one for the assessment of the effectiveness of measures taken to reduce ozone concentrations. To assess the effectiveness of these measures a Level II approach is necessary. This Level II approach should consider the spatio-temporal differences in sensitivity of plants to ozone.

The long-term goal of a Level II approach should be the development of a flux-model (UN ECE 1999). But there are some difficulties to realize a flux based model on a larger scale. Modification of the Level I AOT40 values by using modifying factors could be reasonable as a kind of interim solution. According to UN ECE (1999) the following modifying factors should be considered:

- plant species
- phenology
- soil moisture
- irrigation
- vapor pressure deficit
- temperature
- wind speed
- global radiation
- synergistic effects with other pollutants

Only the modifying factor with the most serious effect on crop loss should be considered (UN ECE 1999). Modifications should be done to the ozone concentration data and not to the calculated AOT40 values.

First of all the availability of data of modifying factors has to be checked. **Plant species** is a superior modifying factor. Species specific characteristics are important when following a Level II approach. The influence of modifying factors on ozone uptake and effects differs from species to species. Under economical point of view wheat is most important. **Phenology** is one of the most important Level II factors (POSCH & FUHRER 1999). For some plants there are substantial differences in sensitivity to ozone due to different phenological stages. First mapping results of a phenologically weigthed AOT40crops will be discussed below. **Soil moisture** has not yet been integrated as a modifying factor, because there is a lack of data. Though the Deutscher Wetterdienst (DWD) releases a daily map of soil moisture, which is based on model data in the internet. But to integrate soil moisture into Level II mapping procedure a hourly resolution is needed. At that time there is no suitable data on soil moisture available. The influence of i**rrigation** is of minor importance in Germany, in particular when looking at crop losses of wheat. There are some regions where plants, especially vegetables are irrigated regularly, but for wheat irrigation is of almost no importance in Germany. **Vapor pressure deficit** can be calculated with the Magnus formula out of data on actual temperature and temperature of the dewpoint. Monitoring data of actual temperature and temperature of the dewpoint are available at Institute of Navigation, Stuttgart University for the years 1994-1998. To use vapor pressure deficit as a modifying factor the influence of vapor pressure deficit on crop loss has to be determined. There are no functions available yet. For **temperature** it is much the same thing. There are data on temperature (hourly averages) available for the years 1988-1998, but there is no function or factor describing the influence of temperature on crop losses of wheat. Model data of **wind speed** in 6-hourly averages could be provided. More information on the influence of wind speed on crop losses of wheat or ozone fluxes into leaves and needles is needed. There is no hourly data on **global radiation** available. At best monthly data can be achieved. Finally, there is no information on **synergistic effects with other pollutants** available.

6.1 Phenologically weigthed AOT40crops

Phenology is one of the most important Level II factors (POSCH & FUHRER 1999). For some plants there are substantial differences in sensitivity to ozone in different phenological stages. Species specific characteristics have to be considered. With regard to crop losses of wheat ozone concentrations from two months before til one month after the anthesis are most important (SOJA et al 1999). Effects of ozone concentrations in the first month after anthesis are about two to three times higher than in the two months before anthesis. SOJA et al (1999) recommend

for the calculation of phenological and geographical weighted AOT40 values multiplication factors (figure 31). These multiplication factors shall compensate the differences in sensitivity to ozone caused by different phenological stages.

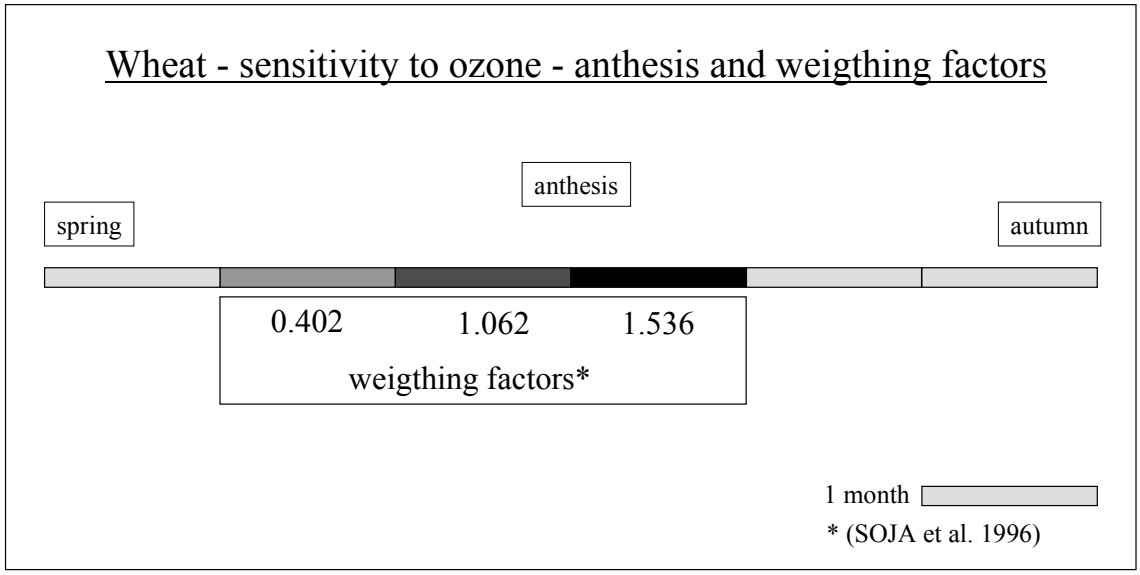


Figure 31: Wheat – sensitivity to ozone – anthesis and weigthing factors.

In Germany there is no data on the beginning of the anthesis of wheat available, for the anthesis of wheat is not observed in IPG's (International Phenological Gardens). Only the beginning of the harvest of wheat is registered there. Winter wheat is harvested six to eight weeks after the beginning of the anthesis. This information could be used to estimate the beginning of the anthesis of wheat on a regional scale. But there is another possibility to estimate the day of the beginning of the anthesis of winter wheat (figure 32). Anthesis of wheat sets in when the so-called GDD (growing degree days) reaches a value of 1150 K-days (Kelvin-days) (POSCH & FUHRER 1999). The GDD or corn growing degree day is an index used to express crop maturity. Growing degree days (GDD) are calculated as the difference between mean daily temperature, and a (base) threshold temperature taken as 0 °C for wheat. The daily differences are summed up to give a cummulative growing degree days for the growing season. A GDD of 1150 has been shown to be a reliable estimator for the anthesis of wheat. This method is proved statistically and now has been applied to the German dataset of 1994.

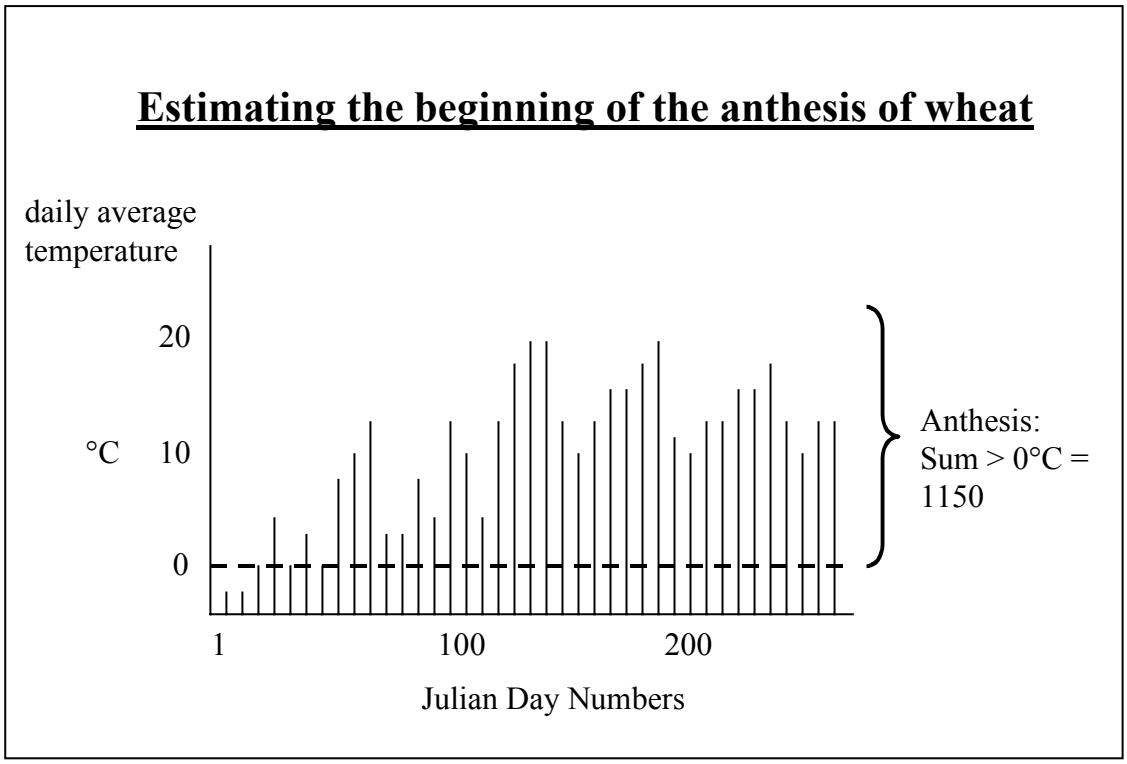


Figure 32: Estimating the beginning of the anthesis of wheat using the growing degree days index.

To estimate the growing degree days data on daily average temperature is needed. In Germany daily average temperature is monitored at about 1000 monitoring sites. The 1994 dataset provides data from 583 monitoring sites. This data has been used to calculate and map the date of the beginning of the anthesis of winter wheat in Germany.

The sum of the daily average temperature above 0 °C is heavily affected by altitude and continentality of the site. In regions favoured by climate anthesis sets in earlier than in high altitudes or regions with oceanic climate. There are considerable differences within Germany. In Level I mapping procedure there has been a fix period of time to be considered to calculate AOT40crops (May – July). Following the modified Level I approach the beginning of the calculation period of AOT40crops can be determined in a site specific way (figure 33).

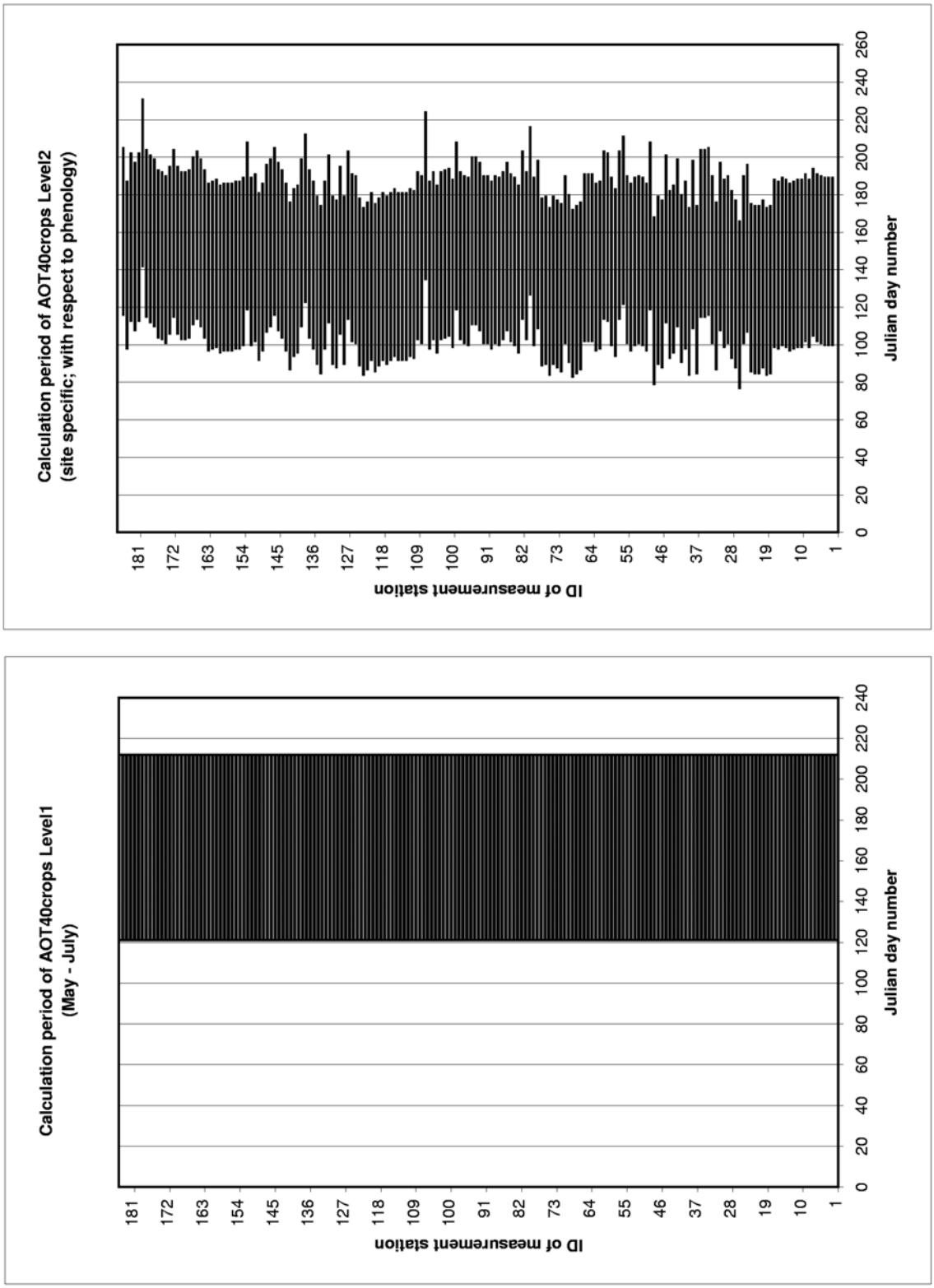


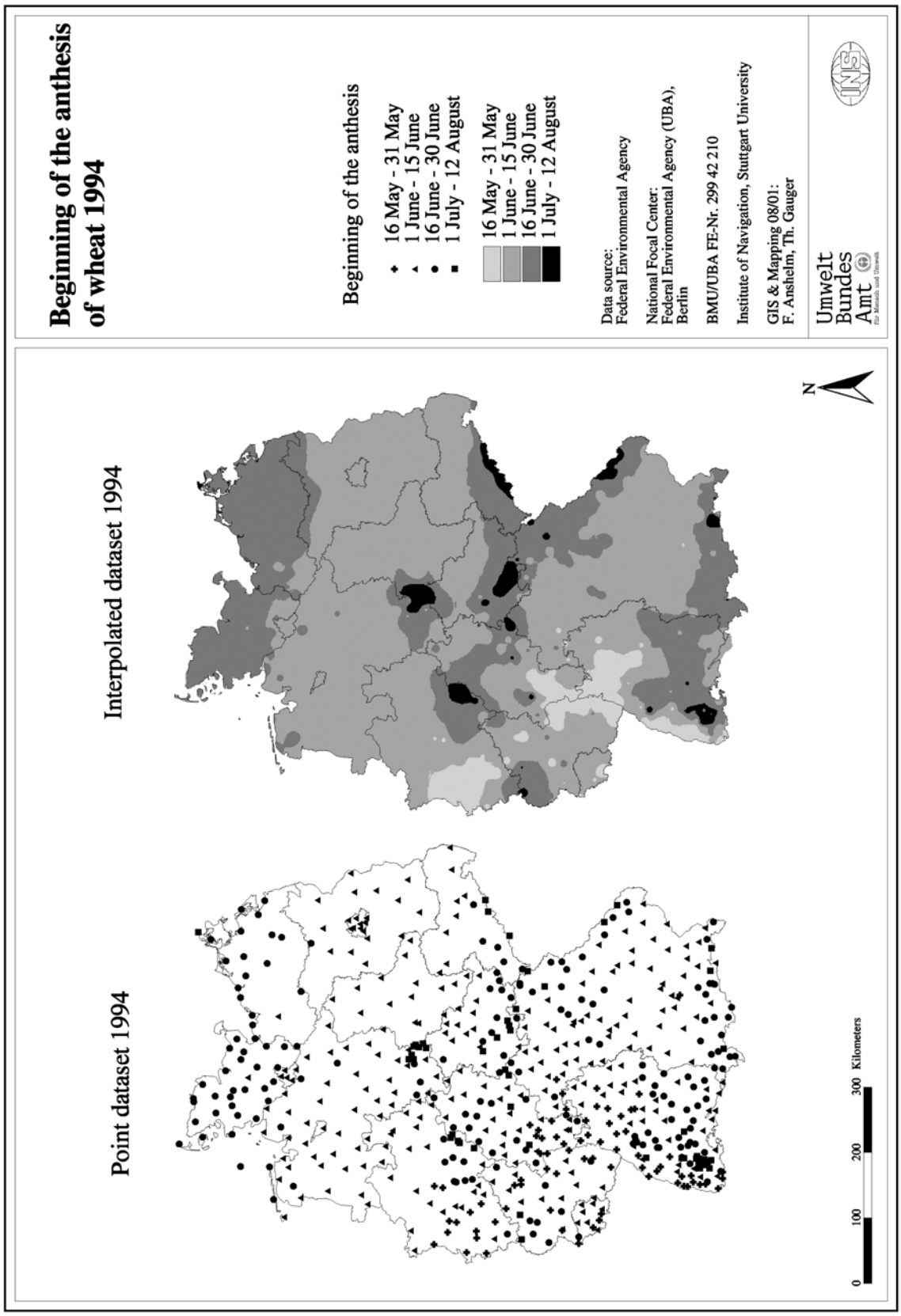
Figure 33: Calculation period of Level I AOT40crops and phenologically weigthed AOT40 crops.

For most of the sites the period for calculating AOT40crops will start earlier. Only for five of 184 monitoring sites the calculation period starts later. These five monitoring sites are situated in high altitudes (> 500 m NN). At 179 monitoring sites the calculation period will start earlier when following the modified Level I approach. Anthesis of wheat sets in earliest at Kehl monitoring site (Julian day number 137 = 17th of May). At Schmücke monitoring site anthesis sets in lastly (Julian day number 202 = 21st of July). On average, anthesis of wheat starts at 9th of June (Julian day number 160). This means the calculation period to be considered for calculation of AOT40crops starts on average three weeks earlier than in Level I mapping procedure.

Map 27 illustrates the beginning of the anthesis of wheat in 1994. In regions favoured by climate the anthesis started between the 16th of May and the end of May (Oberrheinische Tiefebene, Mittelrheintal, Gäulandschaften). In most parts of Germany anthesis started between the 1st and the 15th of June. Near the coast of the North Sea and Baltic Sea as well as in low mountain ranges anthesis started between the 16th of June and the end of June. Finally, there are some regions where anthesis started not before the 1st of July. In high altitudes anthesis started not until the first half of August (The Alps, Southern Black Forest, Harz, Erzgebirge, Thuringian Forest, Bavarian Forest, Rothaargebirge). But in these high altitude regions wheat is not cultivated, because wheat needs at least an annual average temperature of 5 °C to sprout.

The phenologically weighted AOT40crops still is calculated within a three months period. The beginning of this period depends as shown before on the beginning of the anthesis. Ozone concentration data of the month two months before anthesis has to be multiplied by 0.402, data of the last month before anthesis has to be multiplied by 1.062 and the data of the first month after anthesis by 1.536 (figure 31).

Table 5 illustrates results of a comparison of Level I AOT40crops values and phenologically weigthed AOT40crops values (modified Level I approach). The comparison is based on the 1994 data of 183 German monitoring sites. At 56 monitoring sites the phenologically weigthed AOT40crops is less than Level I AOT40crops, at 127 monitoring sites it is higher than Level I AOT40crops. Minimum, maximum, mean and median of the phenologically weighted AOT40crops are higher than Level I AOT40crops values.

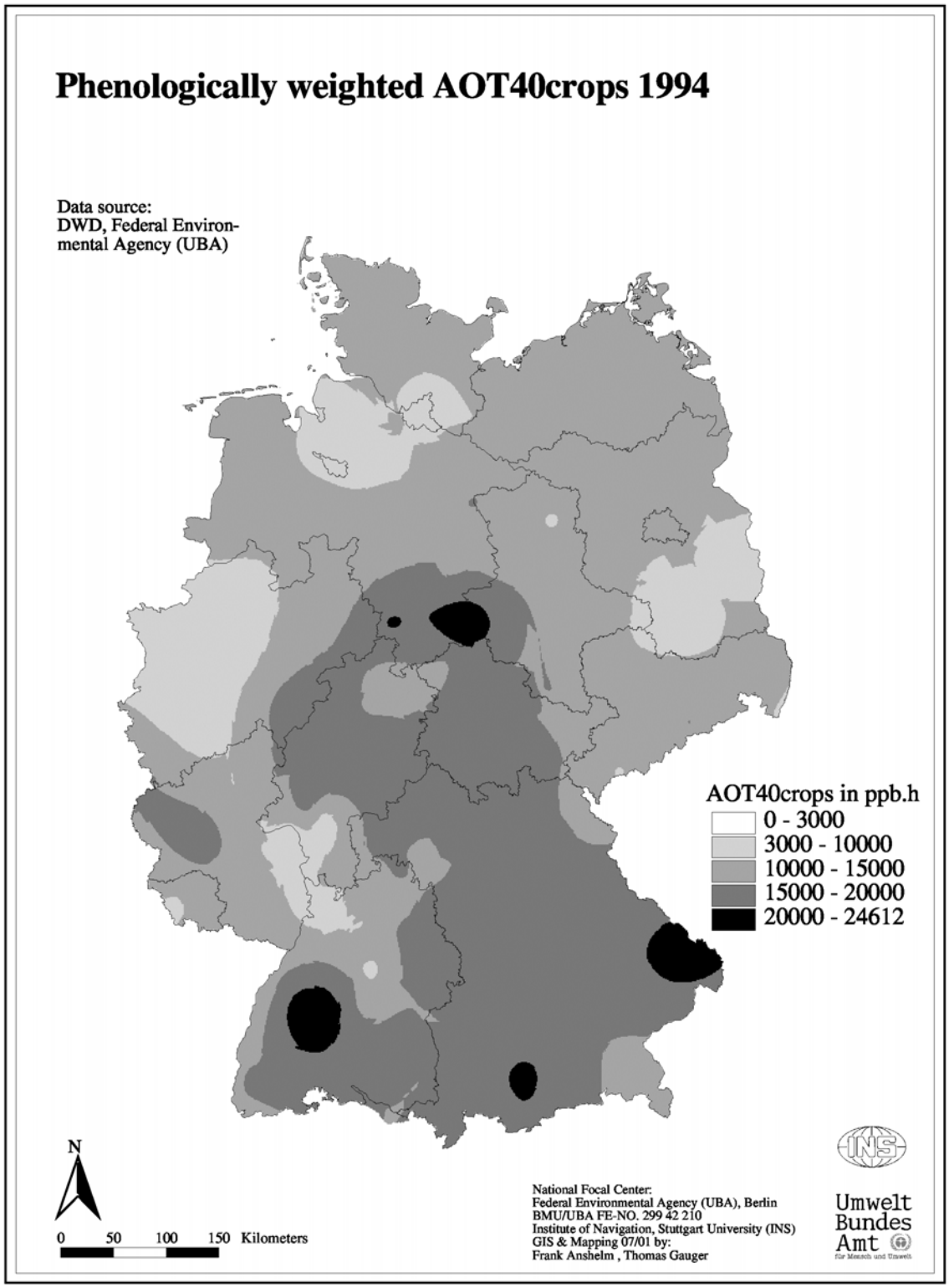


Map 27: Beginning of the anthesis of wheat 1994.

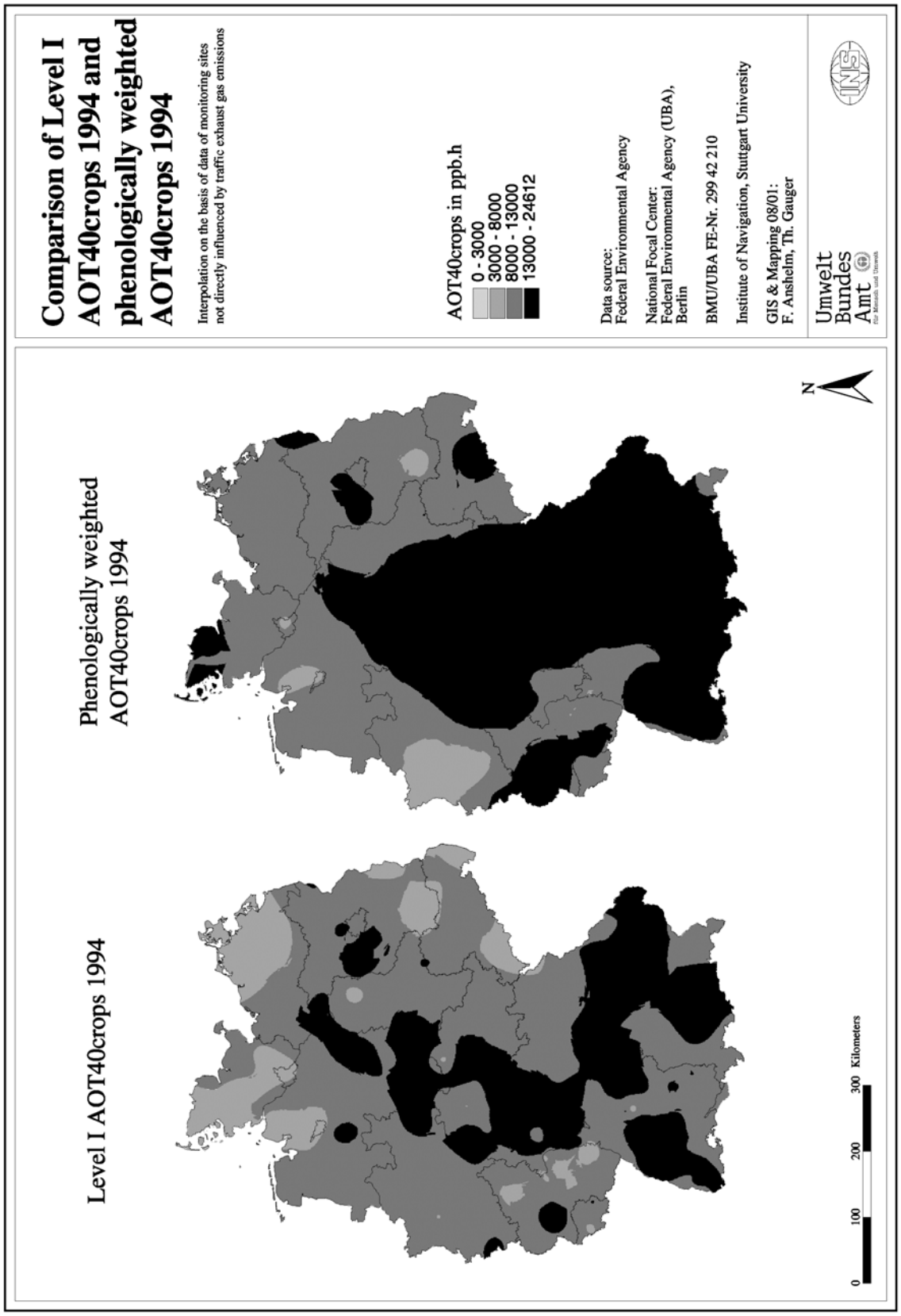
Table 5: Comparison of AOT40crops and phenologically weighted AOT40crops 1994.

Comparison of AOT40crops and phenologically weigthed AOT40crops 1994				
	AOT40crops	mod. AOT40crops	increase in ppb.h	increase in %
min	2712	3001	289	11
max	22472	30992	8520	38
mean	11226	12540	1314	12
median	10999	11843	845	8

Map 28 and 29 illustrate mapping results of phenologically weighted AOT40crops and a comparison of Level I AOT40crops and phenologically weigthed AOT40crops. The Critical Level of 3000 ppb.h is exceeded throughout Germany. In Level I mapping there are a few gridcells near the city of Bremerhaven where the Critical Level is not exceeded. The area with AOT40crops of more than 13000 ppb.h is much bigger in phenologically weigthed mapping. But there also some regions where the AOT40crops is getting lower when considering phenology (western North Rhine-Westphalia).



Map 28: Phenologically weighted AOT40crops 1994.



Map 29: Comparison of Level I AOT40crops and phenologically weighted AOT40crops 1994.

7 Summary

Within the research project „Mapping of ecosystem specific long term trends in deposition loads and concentrations of air pollutants in Germany and their comparison with Critical Loads and Critical Levels „ (BMU/UBA FE-NO. 299 42 210) national maps describing concentration of air pollutants and exceedances of Critical Levels have been calculated. Special interest has been put on the detection of long term trends.

Air concentration of SO_2 and exceedances of SO_2 Critical Levels have been calculated and mapped for the years 1985-1999. Since 1987, there is a downward trend of the annual average concentration of SO_2 within Germany. This applies to exceedances of SO_2 Critical Levels, too. Since 1997, there is no more exceedance of the SO_2 Critical Level for crops (annual average: 30 µg/m³) throughout Germany. The SO_2 Critical Level for forest ecosystems and natural vegetation (annual average: 20 µg/m³) has not been exceeded in 1998 and 1999. There still have been some slight exceedances of the SO_2 Critical Level for lichens (annual average: 10 µg/m³) in 1999, but this on a very small area and there is a stable downward trend.

Air concentration of NO_2 has been calculated and mapped for the years 1985-1999. There is a slight downward trend of the annual average concentrations. The next years will show, whether there is a reliable trend in NO_2 concenteation data. Data of the concentration of NO is available for the years 1992-1999, but mapping is possible for the years 1995-1999 only. NO data of the years before 1995 are too patchy for mapping on the national scale. The series of NO_x concentration maps ($NO_x = NO_2 + NO$) and exceedance of NO_x Critical Levels therefore is very short (1995-1999). There is no reliable trend detectable. The NO_x Critical Level for Vegetation (annual average: 30 µg/m³) has been exceeded in some parts of Germany during 1995 and 1999. There are exceedances up to more than 100 % in every year, but there are regions of no exceedance, too.

Air concentration of ozone has been calculated and mapped for the years 1990-1999. There is no clear trend in annual average ozone concentration data. The ozone Critical Level for crops (AOT40crops: 3000 ppb.h) has been exceeded almost throughout Germany in the whole 1990ies. There are AOT40crops values up to more than 20000 ppb.h. Due to the weather conditions in summer time, there are some years with high exceedances (1992, 1994, 1995) and some years with relatively low exceedances (1990, 1991, 1997) of AOT40crops. Looking at the ten years average AOT40crops (1990-1999) values there are some regions with relatively low ex

ceedances (northern parts of Germany) and some regions with high exceedances (south-western parts of Germany). The ozone Critical Level for forests (AOT40forests: 10000 ppb.h) has been exceeded throughout the whole 1990ies too, but there are bigger regions, where the Critical Level has not been exceeded. There are AOT40forests values up to more than 40000 ppb.h.. Highest AOT40forests values can be stated in the south-western parts of Germany and in areas of high elevation. Looking at the ten years average AOT40forests (1990-1999) values there are some regions, mainly in the northern and north-western parts of Germany, where the Critical Level for forest ecosystems is not exceeded.

Ozone and health effects has been another important topic within this project. AOT60 and exceedance of the WHO Guideline Level for the protection of human health (WHO120) has been calculated and mapped for the years 1990-1999. There are AOT60 values up to more than 30000 ppb.h. WHO Guideline Level for the protection of human health has been exceeded in all years. In some years and regions there are more than 100 days of exceedance of the WHO Guideline Level. But there are also regions, where the WHO Guideline Level (max. 20 days of exceedance averaged over three years) has not been exceeded during the 1990ies.

In the future ozone Level II mapping will become more important (ozone flux model). Modifying AOT40crops by including factors, which can have an influence on ozone uptake into plants, could be a kind of interim solution. This has been shown for phenology, which is one of these so-called Level II factors. Compared to Level I AOT40crops mapping results the phenologically weigthed AOT40crops values has been shown to be higher in most places within Germany. For most of the other potential Level II factors like global radiation, soil moisture, irrigation or vapor pressure deficit there is a lack of data, which aggravates inclusion of these factors into Level I mapping procedure. On the long run a flux based model should be the final goal.

8 Zusammenfassung

Im Rahmen des Forschungsvorhabens „Kartierung ökosystembezogener Langzeittrends atmosphärischer Stoffeinträge und Luftschadstoffkonzentrationen in Deutschland und deren Vergleich mit Critical Loads und Critical Levels „ (BMU/UBA FE-Nr. 299 42 210) wurden nationale Karten angefertigt, die die Konzentration der Luftschadstoffe Schwefeldioxid (SO_2), Stickstoffdioxid (NO_2), Stickoxide (NO_X) und Ozon (O_3) sowie die Überschreitung der Critical Levels für Schwefeldioxid, Stickoxide und Ozon (AOT40, AOT60) darstellen.

Die Schwefeldioxid-Konzentration und die Überschreitung der Critical Levels für SO_2 wurde für die Jahre 1985-1999 berechnet und kartiert. Seit 1987 ist bei den Jahresmittelwerten ein abnehmender Trend feststellbar. In 1999 lag die SO_2-Immission in ganz Deutschland unter 10 µg/m³. Analog zu den Konzentrationswerten haben die Überschreitungen der Critical Levels seit 1987 beständig abgenommen. In den Jahren 1997-1999 konnte innerhalb Deutschlands keine Überschreitung des Critical Levels für landwirtschaftliche Nutzpflanzen (Jahresmittel: 30 µg/m³) mehr festgestellt werden. Der Critical Level für Waldökosysteme und natürliche Vegetation (Jahresmittel: 20 µg/m³) wurde in den Jahren 1998 und 1999 ebenfalls nirgendwo in Deutschland überschritten. Auch die Überschreitung des Critical Levels für Flechten (Jahresmittel: 10 µg/m³) ist seit 1987 stark zurückgegangen. In 1999 gab es auf nur noch auf kleiner Fläche geringfügige Überschreitungen des Critical Levels.

Die Stickstoffdioxid-Konzentration in Deutschland wurde für die Jahre 1985-1999 berechnet und kartiert. Es ist ein sehr schwach abnehmender Trend der Jahresmittelwerte sowie der festgestellten Maximalwerte zu verzeichnen. Ob dies ein stabiler Trend ist wird sich in den nächsten Jahren noch zeigen müssen. Daten zur Stickstoffmonoxid-Konzentration sind für die Jahre 1992-1999 verfügbar. Kartiert werden konnten jedoch lediglich die Jahre 1995-1999. In den Jahren vor 1995 sind die Datensätze zu lückenhaft als dass eine Kartierung im nationalen Maßstab möglich wäre. Aus diesem Grunde ist auch die Kartierung der NO_X-Konzentration (NO_X = NO_2 + NO) und die Kartierung der Überschreitung des Critical Levels für NO_X nur für die Jahre 1995-1999 möglich. Auf Basis der nur fünfjährigen Datenreihe ist keine verlässliche Aussage über einen Trend möglich. Für Stickoxide (NO_X) gibt es nur einen Critical Level für die gesamte Vegetation. Dieser liegt bei einem Jahresmittel der NO_X-Konzentration von 30 µg/m³. Dieser Critical Level wurde zwischen 1995 und 1999 immer wieder und in unterschiedlich starkem Ausmaß überschritten. Es gab Überschreitungen von bis zu mehr als 100 %, aber auch Gebiete, in denen zwischen 1995 und 1999 keine Überschreitung des Critical Levels aufgetreten ist.

Die Jahresmittel der Ozon-Konzentration konnten für die Jahre 1990-1999 berechnet und kartiert werden. Es konnte kein verläßlicher Trend festgestellt werden. Der Critical Level für landwirtschaftliche Nutzpflanzen (AOT40crops: 3000 ppb.h) wurde zwischen 1990 und 1999 in allen Jahren nahezu flächendeckend überschritten. Lediglich im Jahr 1991 gab es im Nordwesten Deutschlands ein großes zusammenhängendes Gebiet, in welchem der Critical Level nicht überschritten wurde. In allen anderen Jahren blieben Unterschreitungen auf kleine Flächen begrenzt. In einzelnen Jahren wurden Werte von mehr als 20000 ppb.h erreicht. Es gibt einzelne Jahre mit sehr hohen Überschreitungen (1992, 1994, 1995) und einige Jahre mit verhätnismäßig geringen Überschreitungen (1990, 1991, 1997). Ursache hierfür sind in erster Linie die unterschiedlichen Wettterbedingungen in den jeweiligen Jahren. Betrachtet man den Mittelwert des 10-Jahreszeitraums 1990-1999 so gibt es Regionen mit relativ geringen Überschreitungen (Norddeutschland) und Gebiete mit verhältnismäßig hohen Überschreitungen (Südwestdeutschland).

Der Critical Level für Waldökosysteme (AOT40forests: 10000 ppb.h) wurde in den 90er Jahren zumeist überschritten. Die Gebiete, in denen der Critical Level nicht überschritten wurde sind etwas größer als bei AOT40crops. Es konnten AOT40forests-Werte von bis zu 40000 ppb.h festgestellt werden. Die höchsten Werte sind an einigen Höhenstationen im Südwesten Deutschlands aufgetreten. Über einen Zeitraum von 10 Jahren gemittelt (1990-1999) werden im Norden Deutschlands sowie im Westen größere zusammenhängende Gebiete erkennbar, in denen der Critical Level für Waldökosysteme nicht überschritten wurde.

Die Wirkung von Ozon auf die menschliche Gesundheit ist ein weiteres wichtiges Thema. Für die Jahre 1990-1999 wurde der AOT60 sowie die Überschreitung des WHO-Leitwerts zum Schutz der menschlichen Gesundheit vor zu hoher Belastung mit bodennahem Ozon berechnet und kartiert. In einzelnen Jahren und Regionen wurden AOT60-Werte von mehr als 30000 ppb.h erreicht. Der WHO-Leitwert zum Schutz der menschlichen Gesundheit wurde in einzelnen Jahren und Regionen an mehr als 100 Tagen überschritten. Die WHO empfiehlt, dass innerhalb eines 3-Jahreszeitraums der Leitwert an nicht mehr als 20 Tagen überschritten werden sollte. In den 3-Jahresmitteln 1991-93, 1994-96 und 1997-99 treten jeweils Überschreitungen von mehr als 60 Tagen auf. Es gibt allerdings auch relativ große Gebiete, in denen der WHO-Leitwert zum Schutz der menschlichen Gesundheit eingehalten wurde. Wie bei allen Bewertungsmaßstäben, die den Parameter Ozon betreffen, treten auch hier starke Schwankungen zwischen den einzelnen Jahren auf – entsprechend den regional oft recht unterschiedlichen Wetterbedingungen im Sommer des jeweiligen Jahres.

Zukünftig wird das bisher angewandte Kartierungsverfahren (Level-I-Ansatz) durch ein Flüsse-Modell abgelöst werden (Level-II-Ansatz). Parameter, die den Ozon-Fluss in die Pflanzen beeinflussen sollen stärker berücksichtigt werden. In einer Übergangsphase – bisher ist eine Modellierung der Ozon-Flüsse im nationalen Maßstab noch mit zu großen Unsicherheiten behaftet – kann die Einbeziehung modifizierender Faktoren in das Level-I-Kartierungsverfahren eine Übergangslösung darstellen. Ein wichtiger, die Ozonaufnahme der Pflanzen beeinflussender Parameter ist die Phänologie der Pflanzen. Pflanzen sind während unterschiedlicher phänologischer Stadien unterschiedlich empfindlich gegenüber der Immission von Ozon. Die Phänologie wurde als ein erster modifizierender Faktor in das Level-I-Kartierungsverfahren für AOT40crops integriert. Verglichen mit den Level-I-Kartierungsergebnissen für AOT40crops sind die phänologisch gewichteten Kartierungsergebnisse in den meisten Gebieten Deutschlands etwas höher. Andere potentielle modifizierende Faktoren wie Globalstrahlung, Bodenfeuchte, Bewässerung oder das Dampfdrucksättigungsdefizit der Luft konnten bisher nicht in das Level-I-Kartierungsverfahren integriert werden. Die Zukunft jedoch gehört der Modellierung der Ozon-Flüsse in die Pflanzen (Level-II-Ansatz).

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Annex 1

Types of monitoring stations and list of codes for description of monitoring sites.

Type of station

Rural: station is located outside built-up areas of cities or conurbationd. It is used to monitor ‚regional backgound' air pollution levels. Station is located away from emission sources.

Urban: station used to monitor the ‚average' air pollution levels in urban area (urban background concentration) resulting from transport from outside and from emissions in the city itself. The station is not directly influenced by emission sources like traffic or industry.

Street: station used to monitor the air pollution levels in traffic street with more than 2000 vehicles/day within 50 meters. It is directly and predominantly influenced by traffic emissions. This station is located in urban area but can also be operated along an highway.

Immediate environment codes (as defined in GIRAFE[6])

The immediate environment (within a radius of 0 to 100 m) is described with up to four of the following codes:

large street heavy traffic	I1a
large street medium traffic	I1b
large street low traffic	I1c
large street pedestrian zone	I1d
small street heavy traffic	I2a
small street medium traffic	I2b
small street low traffic	I2c
small street pedestrian zone	I2d
canyon street heavy traffic	I3a
canyon street medium traffic	I3b
canyon street low traffic	I3c
canyon street pedestrian zone	I3d

[6] EU Member States have exchanged information and data from air quality networks and individual stations since 1976. This exchange was based on several Council Decisions (Decision 82/459; 97/101/EC). Technical information on the monitoring stations and a description of the station surroundings are stored in a database called GIRAFE.

footway	I4
front of building	I5
terrace, bell tower,belfry	I6
interior court,school, hospital	I7
trees	I8
large flat area	I9
channel	I91
meadow, field	I92
other	I99

Local environment codes (as defined in GIRAFE)

The local environment (within a radius of 100 m to some km) is described with up to four of the following codes:

urban commercial	Laa
urban industrial	Lab
urban residential	Lac
mixture of commercial, industrial and residential	Lad
industrial heavy concentration	Lba
industrial medium concentration	Lbb
industrial light concentration	Lbc
road traffic heavy	Lca
road traffic medium	Lcb
road traffic light	LCc
commercial	LD
residential (isolated houses)	LE
harbour	LF
airport	LG
park, forest, natural field	LH
agricultural area	LI
mountains, vallexs	LJ
sea side or lake side	LK
other	LL