

Air pollution by ozone in Europe in 1997 and summer 1998

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Preface

This report is based upon data submitted to the European Commission under the Directive 92/72/EEC on air pollution by ozone. The Commission has requested the European Environment Agency (EEA) to assist in this annual reporting by EU Member States since 1995. The voluntary submission to the EEA of information on ozone levels by other European countries made it possible to present an ozone assessment on a wider European scale. The data collection in Phare countries was greatly facilitated by the Phare Topic Link on Air Quality working as part of the European Topic Centre on Air Quality. Two separate reports, one covering the year 1997, the other covering summer 1998, were originally produced by the European Topic Centre on Air Quality and presented to the Member States in their meeting of November 1998. The separate reports are available in electronic form on the EEA homepage on the Internet (<http://eea.eu.int>).

In this EEA Topic Report, the annual 1997 and summer 1998 reports are reproduced in their original form as presented to the Commission. The differences in available data, the set of threshold values, the number of stations, the location of stations and the status of the information (based on validated 1997 data and non-validated 1998 data) justifies such an approach.

A major asset of this report is the timeliness of its delivery. The assessment of ozone episodes in 1998 was based upon data measured only two months earlier, while the normal production time from field measurements to validated assessment reports is rarely less than eighteen months. This timely reporting has only been possible with the support of the individual contact points within each country and the efficient communication established.

The harmful effects of tropospheric ozone on human health and well-being as well as damage to ecosystems is now being recognised as a major concern throughout the European Union. The European Community has taken steps to address the problem through Directive 92/72/EEC on ambient ozone, Directive 96/62/EEC on ambient air quality assessment and management (the Framework Directive), and development of an ozone daughter Directive, as well as the decision to develop a Community strategy for the reduction of ozone pollution. The measures necessary to abate pollution remain however a responsibility of each Member State and require political decisions with cost implications and consequences for the development of activities in the society. In this political process objective and reliable information on the extent and severity of the problem is essential.

It is the intention of EEA to continue the yearly reporting and assessment of ground level ozone in Europe in close co-operation with the European Commission, Member States and other countries.

Gordon McInnes
Programme manager

Summary

This report summarises the annual information on ozone monitoring stations and exceedances of ozone threshold values during 1997 and gives a first evaluation of the observed exceedances of the thresholds during Summer 1998 (April-August). According to the Council Directive 92/72/EEC on air pollution by ozone, EU Member States have to provide information on ozone levels (statistical parameters, number and duration of exceedances of specified threshold values) on an annual basis before 1 July of the next year. Additionally, exceedances of the threshold values for population information and warning, as set in the Directive, must be reported to the Commission within one month after occurrence.

The analysis for the year 1997 presented in this report is based on information made available before 12 August 1998. By then, information for the calendar year 1997 was received from all Member States and, on a voluntary basis, from 5 other European countries (Switzerland, Czech Republic, Latvia, Norway and Poland). Information on the situation in Slovakia was received one month later; unfortunately too late to include fully in this report. All information has been submitted in computer readable form.

For the summer 1998-period, all 15 EU Member States provided information on the observed exceedances in time (the deadline for transmitting data was set at 20 September 1998), or indicated that no exceedances were observed. It is greatly appreciated by the Commission that Member States were able to transmit August exceedance data before the formal deadline as set in the Directive. Some countries submitted files which were not formatted according to the prescribed Commission requirements. These files by exception were converted at the European Topic Centre on Air Quality (ETC-AQ) for further processing.

From an evaluation of the exceedances and annual statistics, the following conclusions are drawn:

- In 1997 the threshold value set for the protection of human health ($110 \mu\text{g}/\text{m}^3$ for 8-hourly average concentrations) was exceeded substantially and in all reporting countries: the total number of exceedances reported for stations within in the EU approached 20,000; on the average, this threshold was exceeded per station on more than 23 days per year.
- In 1997 the threshold value of daily average concentrations set for the protection of vegetation ($65 \mu\text{g}/\text{m}^3$) was exceeded substantially (by up to a factor 3), widely (in all reporting countries) and frequently: at 52 stations located in 10 different countries exceedances have been reported during more than 200 days.

- The threshold value of hourly average concentrations set for the protection of vegetation (200 $\mu\text{g}/\text{m}^3$) was exceeded largely and widely (reported by 10 EU Member States and in three out of the six other European countries) on a limited number of days (in total 909 exceedance days were counted at EU monitoring stations).
- In 1997 the threshold value for providing information to the population (180 $\mu\text{g}/\text{m}^3$ for hourly values) was exceeded in 15 countries of which 12 EU Member States during a limited number of days: in EU Member States a total of 2336 exceedance days was counted. During Summer 1998 this threshold was exceeded in all Member States with the exception of Ireland, Finland, Denmark and Sweden. The number of days on which at least one exceedance was observed ranged from 7 in Belgium to 74 in Spain. 61 % of all stations reported one or more exceedance. On average 3.9 exceedances occurred this year at stations which recorded at least one exceedance; the average exceedance duration was 2.7 hours. The average maximum hourly concentration during an exceedance of the threshold this year was 201 $\mu\text{g}/\text{m}^3$.
- Exceedance of the threshold value for warning of the population was reported in 1997 from one station (on 18 June 1997, 13.00 an hourly ozone concentration of 383 $\mu\text{g}/\text{m}^3$ was measured at Lykovrissi, Athens, Greece). The threshold was exceeded on three days during summer 1998 in the Athens conurbation (2 and 3 July 1998 at 2 stations, 29 July 1998 at 1 station) and at 1 station on 7 August 1998 in France.
- Ozone monitoring data for the year 1997 were received from 984 stations within the EU and from 100 stations in other countries; during Summer 1998 a comparable number of stations was operational.
- Spatial coverage and documentation on monitoring data quality need improvement. Depending on the local situation, the ozone monitoring stations are characterised as rural, urban, street or other (e.g. industrial). The present subset of rural stations is not representative for the land area of the EU: the subset is estimated to cover about 40-50% of the area. The geographical coverage of the rural stations is rather adequate in North West Europe but in other regions gaps are noted.
- A limited presentation of the percentile values observed in the period 1989-1997 is given for four Member States for which this information was available. Based on the reported data no firm conclusion concerning a trend in percentile values can be given.

DISCLAIMER

The information presented in part II, *Information document concerning air pollution by ozone*, is partly based on non-validated monitoring data and hence should be regarded as preliminary

PART I

Exceedance of EC ozone threshold values in Europe in 1997

Summary based on the information reported in the framework of
the Council Directive 92/72/EEC on air pollution by ozone

Report to the Commission by the European Environment Agency
European Topic Centre on Air Quality

Frank de Leeuw
Tim de Paus

October 1998

1. Introduction

Ozone, O₃, is a strong photochemical oxidant which causes serious health problems and damage to materials and ecosystems. Human exposure to elevated levels of ozone concentrations can give rise to inflammatory responses and decreases in lung function. Symptoms observed are cough, chest pain, difficulty in breathing, headache and eye irritation. Both laboratory and epidemiological data indicate large variations between individuals in response to episodic ozone exposure, the effects seem to be more pronounced in children than in adults. Studies indicate that exposure to ozone concentrations in the range 160-360 µg/m³ for a period of 1-8 hours - concentrations often observed in ambient air over Europe - reduces various pulmonary functions.

Ozone exposure of ecosystems and agricultural crops results in visible foliar injury and in reductions in crop yield and seed production. For vegetation a long-term, growing season averaged exposure rather than a episodic exposure is of concern. Adverse effects on vegetation can be noted at relatively low ozone levels. Within the framework of the UN-ECE Convention on Long-Range Transboundary Air Pollution the critical level¹ for ozone is expressed as the accumulated ozone exposure above a threshold of 40 ppb (corresponding with 80 µg/m³). Guideline values of this accumulated ozone exposure of 3000 ppb.h and 10,000 ppb.h are given for crops and forest, respectively. The World Health Organization in Europe (WHO) came forward with similar guidelines (WHO, 1996b).

It is known that ozone affects materials such as natural and synthetic rubbers, coatings and textiles. However, there are today serious gaps in knowledge on the mechanisms of damage, the attribution of ozone to damage in comparison to other factors and the economic evaluation of such damage. As far as understood there is no 'no-effect level' of ozone for material corrosion; it is assumed that dose-response relations for materials are linear or nearly linear under ambient conditions. Recently, synergistic effects of ozone in combination with the acidifying components SO₂ and NO₂ have been reported to lead to increased corrosion on building materials like steel, zinc, copper, aluminium and bronze.

In view of the harmful effects of photochemical pollution in the lower levels of the atmosphere, the Council adopted in 1992 the Directive 92/72/EEC on air pollution by ozone. The Directive came into force in March 1994. It established procedures for harmonised monitoring of ozone concentrations, for exchange of information, for communication with and alerting of the population regarding ozone and to optimise the action needed to reduce ozone formation.

¹ Critical levels are defined as concentrations of pollutants in the atmosphere above which direct adverse effects on receptors, such as plants, ecosystems or materials may occur according to present knowledge (Bull, 1991).

Article 6 of the Directive specifies how the information on monitoring results must be provided by the Member States to the Commission. Regarding the time frame, two main types of reporting can be distinguished. Information on exceedances of the so-called information threshold (article 6 sub 2) and warning threshold (article 6 sub 3) for the ozone concentration is to be provided within one month after occurrence. Information on exceedances of all threshold values given in Article 6 must be provided within six months following the annual reference period (article 6 sub 1). Article 7 of the Directive stipulates that the Commission shall at least once a year evaluate the data collected under the Directive. The present report gives an overview of ozone monitoring results of 1997. Similar overviews of the 1994, 1995 and 1996 annual data have been prepared by the European Topic Centre on Air Quality (de Leeuw *et al.*, 1995; 1996; 1997). Prior to the current report an overview on ozone threshold exceedances during Summer 1997 (April-August) has been presented to the Commission (de Leeuw *et al.*, 1997).

Harmful ozone concentrations are observed over the whole of Europe. Formation of ozone takes place at various space and time scales: the high emission density of reactive precursors in urbanised areas might lead to high ozone levels within the city or at short distances downwind. But ozone precursors may also be transported over distances of hundreds to thousands kilometres, resulting in ozone formation far from the sources. For improving the insight in current ambient ozone levels over Europe, countries outside the European Union have been asked by the European Environment Agency (EEA) to provide information on ozone exceedances in line with the Ozone Directive.

The data reported here do not cover all ozone monitoring stations in the European Union. For inclusion in this report, the data must satisfy certain criteria stipulated in the Directive, concerning *inter alia* measuring methods, sampling methods, station siting, quality assurance and documentation. Formats on the transfer of data have been defined by the Expert Group on Photochemical Pollution. This group, established by the Commission following Article 7 of the Directive, had several meetings to coordinate the work within the Member States and the Commission in the framework of the Directive.

Background information on the current experience and knowledge concerning photochemical air pollution, dealing in particular with the phenomenology of ozone, the scientific understanding as based on experiments and theory, and the insights from modelling studies on the relation between ozone levels and precursor emissions, may be found in Borrell and Van den Hout (1995), Derwent and Van den Hout (1995), Barrett and Berge (1996) and in *Tropospheric ozone in the European Union. The consolidated report* (Beck *et al.*, 1998) which is currently in preparation following Article 8 of the Directive. In the process of preparing the new 'Ozone Daughter Directive', a comprehensive review of almost all aspects concerning ozone pollution is currently prepared by an expert group established by the European Commission. This so-called *Position Paper on*

Ozone will be published in 1999. Some aspects of the ozone phenomenology are briefly discussed in Annex II.

In the very near future a new ozone daughter directive will be put forward by the Commission. In this new proposal long-term objectives and target values for the protection of human health and vegetation will be defined based on the revised guidelines of WHO (WHO, 1996). The target values should be attained, as far as possible, in 2010. In addition, the draft ozone directive proposes alert thresholds for informing sensitive and general information. Whereas for humans an exposure period 8 hours is considered, for the protection of vegetation the relevant period extends over the whole growing season (three months). As a rule of thumb one might say that the targets for protection of vegetation will be more difficult to meet than the human health related targets.

While the revised WHO guidelines are also relevant in the context of preparing the new daughter directive for ozone an attempt has been made to evaluate the ozone data submitted under the framework of the current directive against these guideline levels, see section 4.5.

2. Data reporting

2.1. Introduction

According to the Ozone Directive, the EU Member States have to provide the following information for the annual reference period:

- maximum, median and 98 percentile value of 1h- and 8h- average ozone concentrations;
- the number, date and duration of periods during which threshold values as presented in Table 1 are exceeded and the maximum concentrations recorded during each occurrence.

In addition to this annual report, Member States must inform the Commission on a monthly basis in case exceedances of the information and warning threshold values are observed. In this report only data reported on an annual basis will be considered.

Table 1: Threshold values for ozone concentrations (in $\mu\text{g}/\text{m}^3$)

Threshold for:	Concentration	Averaging period
health protection	110	8 h
vegetation protection	200	1 h
	65	24 h
population information	180	1 h
population warning	360	1 h

A group of experts from the Members States have followed the practical implementation of the Directive. Among other items this group discussed procedures for data reporting. The formats for information and data exchanges have been defined in the document 'Council Directive 92/72/EEC on air pollution by ozone. Information and data exchange/formats', Doc.Rev. 11/243/95. In general terms, the requested information consists of two parts:

1. information on stations and measurements techniques (Ozone Directive, article 4.2, indents 1 and 2);
2. information on ozone concentration: annual statistics and threshold exceedances (Ozone Directive, article 6.1).

Based on the experiences in processing the data for the 1994 annual report, the European Topic Centre on Air Quality (ETC-AQ) provided remarks concerning data transmission and suggestions for improvement which were discussed in the Expert Group on Photochemical Pollution. Considering the increasing amount of data requiring processing, as well as the improvement of the transfer of data relating to the implementation of the Directive, the Commission has prepared an update (April 1996) of the data exchange format. The major changes concern the transfer of additional information:

- type of station: definition of the location of stations as recommended in the decision on Exchanges of Information (97/101/EC);
- altitude of stations as recommended by the Expert Group;
- NO_x and VOC data, according to Annex 2.3 of the ozone directive;
- file names: it is recommended to define unique names for all files in order to improve the management and transfer of the data files.

For submission of the 1997 data no further modifications in data requirements and data exchange formats have been made. Non-EU countries have been requested to submit their data in agreement with these data exchange formats.

2.2. Data handling

According to the Ozone Directive the requested information over 1997 should have been forwarded to the Commission before 1 July 1998. All data forwarded by the Commission and received at the European Topic Centre on Air Quality (ETC-AQ) before 12 August 1998 has been processed in this report. In this report the definition of the data formats as described in the document Doc.Rev. 11/243/95 and its April 1996 amendment is used as a reference. If necessary, files were converted at the ETC-AQ for further processing.

Non-EU Member States were contacted by the European Environment Agency, through the Phare Topic Link on Air Quality or, in case of Switzerland, directly and asked to voluntarily submit data following the standard formats on data exchange. Information was received from Switzerland, the Czech Republic, Latvia, Norway and Poland².

The summary of received information presented in Table 2 shows that the information for the EU is nearly complete. However, analysing the data revealed a number of errors, ambiguities or missing data. It was attempted to contact national data suppliers to clarify these points but generally this was not successful. Lack of e-mail addresses or fax-numbers hampered a rapid dialogue.

Spain has not submitted statistical information and only submitted an incomplete set of exceedances.

The data received from Italy was of a low quality. Typical errors frequently found in the Italian information are:

- incomplete set of station information (co-ordinates, unique coding, station type, etc. is missing);
- incomplete date of exceedance (month but not day or hour is given);

² At a very late moment (18 September 1998) data from 8 stations in Slovakia was received. Although the data was delivered according to the agreed formats, the limited time and resources did not allow to fully include the data in this report. Data from Slovakia are included only in the overview tables 2-5 presented in Chapter 2 and 3.

- inconsistent information on exceedances of the 180 $\mu\text{g}/\text{m}^3$ and 200 $\mu\text{g}/\text{m}^3$ threshold: for a number of stations, exceedances of the 200 $\mu\text{g}/\text{m}^3$ threshold are not quoted as exceedance of the 180 $\mu\text{g}/\text{m}^3$ threshold;
- there are clear indications that for a number of stations, in case of exceedance of the daily threshold value of 65 $\mu\text{g}/\text{m}^3$ the quoted maximum concentration refers to an hourly averaged concentration instead of the daily mean.

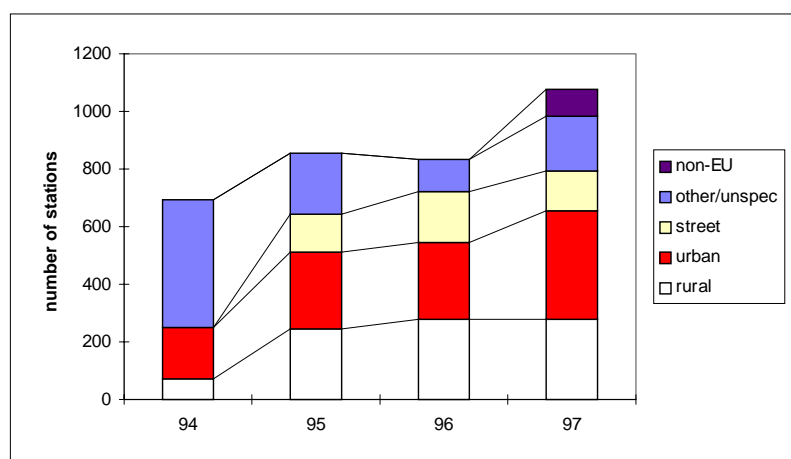
The data from France contain inconsistent information on exceedances of the 180 $\mu\text{g}/\text{m}^3$ and 200 $\mu\text{g}/\text{m}^3$ threshold: for a number of stations, exceedances of the 200 $\mu\text{g}/\text{m}^3$ threshold are not quoted as exceedance of the 180 $\mu\text{g}/\text{m}^3$ threshold.

The statistical information of the Netherlands was in an incorrect format; information on data coverage was not given. This precluded the further use of the Dutch statistical information in the analysis.

A *measurement techniques file* was not received from a number of countries. Voluntary data on precursor concentrations was received for nine Member States and two additional countries.

For 1997 information on ozone concentrations (annual statistics and/or exceedance information) was received for 1076 monitoring stations of which 984 were located in EU Member States. Figure 1 indicates that the number of reporting stations is still growing. The large increase compared to 1996 is mainly due to the fact that over 1996 only 13 Member States submitted information.

Figure 1: Number of stations reporting ozone data within the framework of the ozone directive.



Note that 1996 refers to the actual reporting Member States. For stations in non-EU countries a station classification is known but not resolved in this figure to maintain consistency with earlier years for EU Member States.

Table 2: Summary of information received for 1997

country	station info	meas. tech	statistics	exceedance	NO ₂	NO _x	VOC
AT	✓	✓	✓	✓	✗	✗	✗
BE	✓	✗	✓	✓	✓	✗	✓
DE	✓	✗	✓	✓	✗	✗	✗
DK	✓	✓	✓	✓	✓	✓	✗
ES	✓ ⁽⁴⁾	✓ ⁽⁴⁾	✗	✓ ⁽³⁾	✗	✗	✗
FI	✓	✓	✓	✓	✓	✓	✗
FR	✓	✓	✓	✓	✗	✗	✗
GB	✓	✓	✓	✓	✓	✓	✗
GR	✓	✓	✓	✓	✓	✓ ⁽⁶⁾	✗
IE	✓	✓	✓	✓	✗	✗	✗
IT	✓	✓	✓	✓	✓	✓	✓
LU	✓	✓	✓	✓	✓	✓	✓
NL	✓	✗	✓ ⁽¹⁾	✓ ⁽²⁾	✓ ⁽¹⁾	✓ ⁽¹⁾	✗
PT	✓	✓	✓	✓	✓	✓	✗
SE	✓	✓	✓	✓	✗	✗	✗
CH	✓	✓	✓	✓	✓	✓	✗
CZ	✓	✓	✓	✓	✓	✓	✓
LV	✗	✗	✓ ⁽⁵⁾	✓ ⁽⁵⁾	✗	✗	✗
NO	✓	✗	✓	✓	✗	✗	✗
PO	✓	✗	✓	✓	✗	✗	✗
SK	✓	✓	✓	✓	✗	✗	✗

✓: delivered;

✗: not delivered;

(1) wrong format;

(2) originally data in wrong format; updated file received 18 August 1998;

(3) only period March-October; only exceedances of 180 µg/m³ (1h-averaged);

(4) no 1997-update received; information submitted for 1996 has been used;

(5) hourly data submitted for period April-December 1997; exceedances and statistics calculated by ETC/AQ;

(6) Information on NO₂ and NO (not NO_x) has been received.

3. Survey of reported data for 1997

The location of monitoring stations in EU Member States which are used for the implementation of the Ozone Directive and which are reporting over 1997, is presented in Map 1. Stations located in other European countries which have reported ozone data on a voluntary basis are shown as well. In total information for 984 stations in 15 Member States has been received; for 100 stations in the six reporting non-EU countries information has been received. All reporting countries use the reference method (UV absorption) as prescribed in Annex V of the Ozone Directive. However, at a limited number of stations chemiluminescence is used.

In 1997 only one exceedance of the threshold value of $360 \mu\text{g}/\text{m}^3$ for hourly values has been reported³: on 18 June, 13.00 a hourly ozone concentration of $383 \mu\text{g}/\text{m}^3$ was measured at Lykovrissi (Athens, Greece). This day was the start of an ozone episode in Athens. The station Lykovrissi reported exceedances of the human health related threshold value of $110 \mu\text{g}/\text{m}^3$ for 8-hourly averaged during the period 18-24 June. During the whole month of June, concentrations were high in Athens: on 17 days at least one (out of seven) stations reported an hourly value exceeding $180 \mu\text{g}/\text{m}^3$. On 22 June all seven stations reported an exceedance with an maximum of $229 \mu\text{g}/\text{m}^3$. For a more extensive description of this episode see de Leeuw *et al.* (1997).

A summary of the maximum concentration measured at any of the reporting stations where exceedance⁴ of a threshold value is observed is presented in Table 3. When no exceedances of a threshold have been reported by a country this is indicated with a dash (-).

³ On 20 June 1997 the Italian station *Via Venezia* in Caltanissetta (Sicilia) reported an exceedance of the threshold value of $200 \mu\text{g}/\text{m}^3$ during two hours; as maximum concentration during this exceedance a value of $379 \mu\text{g}/\text{m}^3$ was reported. This value has not been reported as an exceedance of the $360 \mu\text{g}/\text{m}^3$ level. According to information from the Italian data supplier, this value is in error and should be disregarded; another exceedance, observed at nighttime in the winter at the same station, should be disregarded as well (Mr. P. Lagrotta, personal communication).

⁴ In this report exceedances are counted on a daily basis, that is, a day on which at least one 1h- or 8h-concentration exceeds the threshold value, is marked as an exceedance.

Map 1: Location of ozone monitoring stations as reported by Member States in the framework of the Ozone Directive for the reference period 1997. Stations for which other European countries submitted information are shown as well.

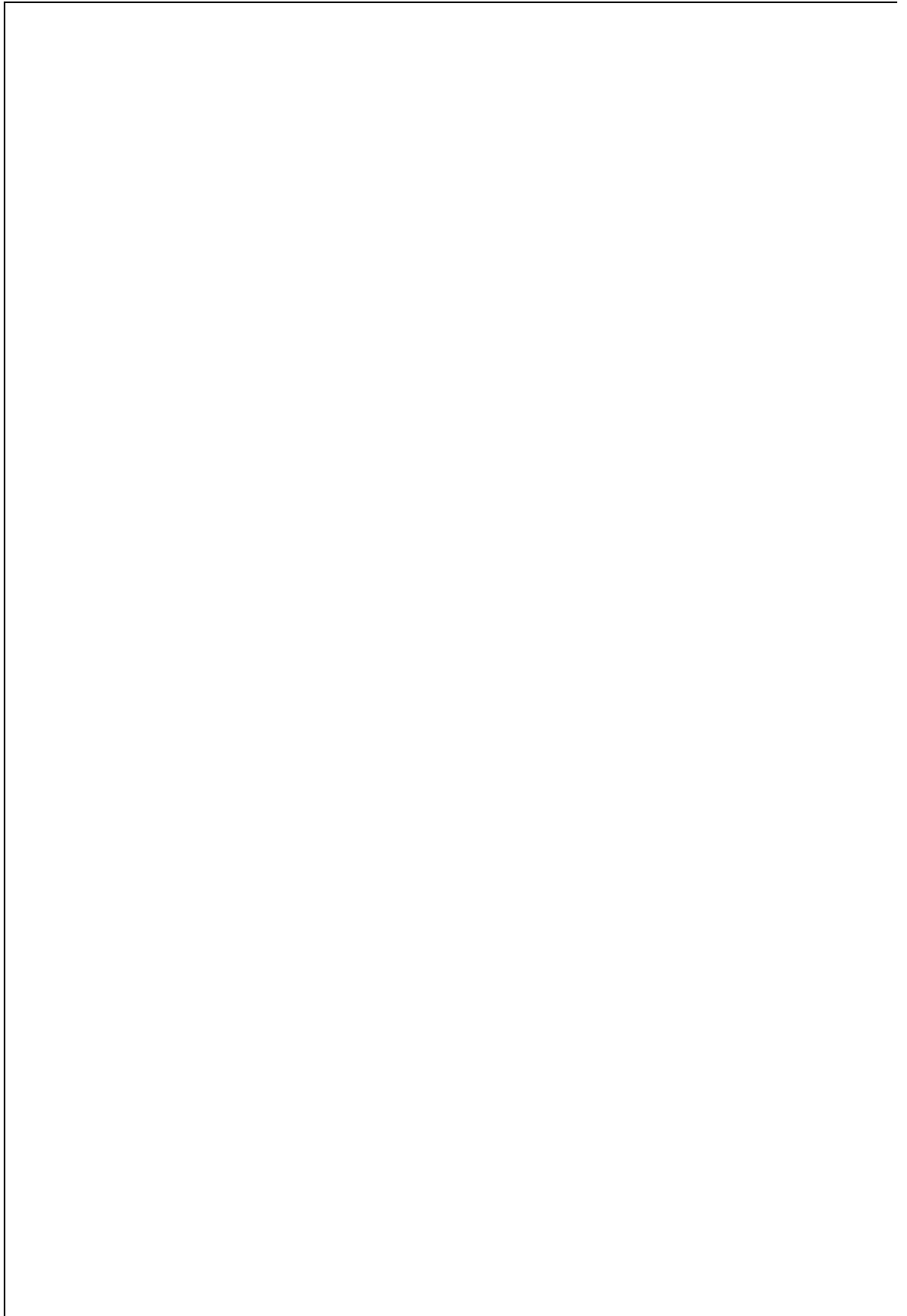


Table 3: Maximum ozone concentrations (in $\mu\text{g}/\text{m}^3$) measured during a period of exceedance of threshold values (reference period 1 January - 31 December 1997). A dash (—) indicates that no exceedances have been observed; na indicates that no data are available.

	180 (1h)	200 (1h)	360 (1h)	110 (8h ^(a))	110 (8h ^(b))	65 (24h)
AT	228	228	—	166	171	153
BE	277	277	—	205	219	171
DE	253	253	—	221	215	214
DK	181	—	—	141	155	133
ES	279	279 ⁽¹⁾	—	na	na	na
FI	—	—	—	147	136	119
FR	199 ⁽²⁾	270	—	184	210	170
GB	314	314	—	188	195	140
GR	383	383	383	221	246	138
IE	—	—	—	151	131	133
IT	353	353	—	207	230	353 ⁽³⁾
LU	203	203	—	166	174	148
NL	266	266	—	183	199	121
PT	271	271	—	204	230	174
SE	—	—	—	141	144	128
EU-15	383	383	383	221	246	353 ⁽³⁾
CH	274	274	—	182	208	166
CZ	200	200	—	177	182	148
LV	—	—	—	115	127	102
NO	—	—	—	162	162	162
PL	222	222	—	163	168	135
SK	—	—	—	135	149	122
all	383	383	383	221	246	353 ⁽³⁾

(a) based on three non-overlapping eight hourly values between 0.00-8.00; 8.00-16.00; 16.00-24.00;

(b) based on the eight hourly value between 12.00 - 20.00;

(1) exceedances of the threshold of 200 $\mu\text{g}/\text{m}^3$ have not been reported; a maximum concentration of 279 $\mu\text{g}/\text{m}^3$ is deduced from the information on exceedances of the threshold of 180 $\mu\text{g}/\text{m}^3$;

(2) for a number of stations exceedances of the 200 $\mu\text{g}/\text{m}^3$ threshold have not been reported as exceedances of the 180 $\mu\text{g}/\text{m}^3$ threshold;

(3) there are clear indications that the maximum concentration refers to an hourly, not daily averaged value.

As the number of monitoring stations differs widely from country to country, the absolute number of exceedances is less suitable for comparison. Therefore, the concept of 'occurrence of exceedances' is introduced here. Occurrence of exceedances is defined as the average number of observed exceedances per country, that is, the total number of exceedances summed over all the stations of a country divided by the total number of reporting stations. A summary of occurrence of exceedances is presented in Table 4. Still, Table 4 has to be interpreted carefully as there are additional reasons which hamper a comparison between the countries. Firstly, the local environment (in particular NO_x sources) influence the ozone levels; the differences between countries partly result from the differences in the ratios of street, urban and rural stations. Secondly, about 20% of the reporting stations show a data coverage of less than 75% (or data coverage is not known); this might result in an underestimation of the

number of exceedances. Also the differences in definition of an exceedance (some countries count an exceedance when the concentration is GREATER THAN the threshold value, other countries count it when the concentrations is EQUAL TO OR GREATER THAN the threshold) may introduce difference in occurrence of exceedances.

Adverse effects of ozone on human health and vegetation will not only depend on the frequency by which a threshold is violated but also on the severity of the exceedance. The severity of an exceedance can be expressed by the *Accumulated exposure Over a Threshold (AOT)*. AOT-values are calculated on basis on hourly concentration according to:

$$AOT = \sum_i^N (C_i - T) \cdot \delta_i$$

where $\delta_i = 0$ for concentration C_i less than threshold T and $\delta_i = 1$ when C_i exceeds the threshold T ; N is the number of valid measurements. The AOT is expressed in *concentration x time* units, for example in *ppm.h* or *mg.h.m³*. From the received information it is not possible to calculate an AOT-level as during a period of exceedance only the maximum concentration is given. As an approximation of the exposure we can calculate an *exceedance rate* which is defined as:

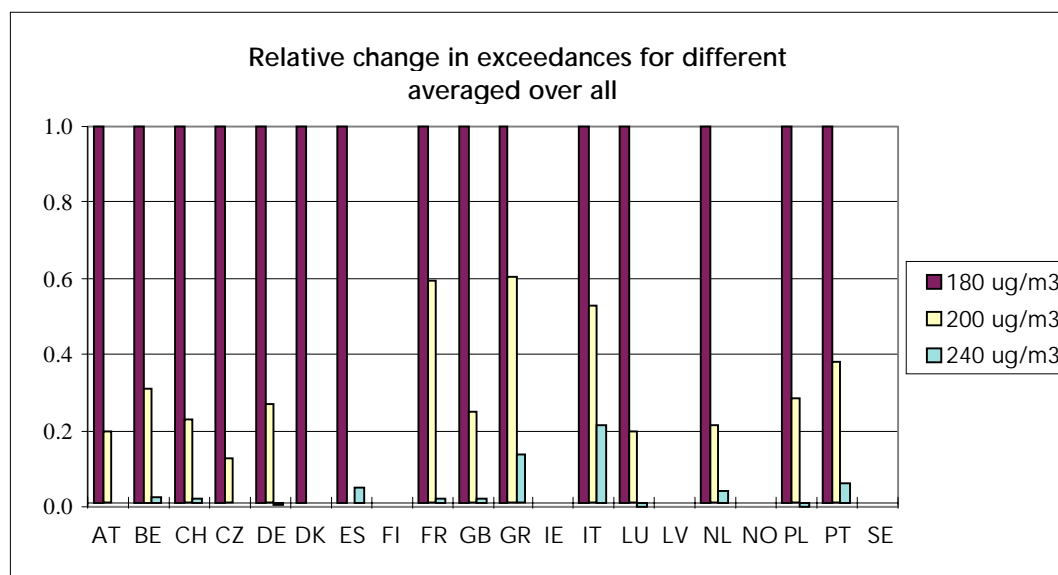
$$ER = \sum_i^{NET} (C_{i,max} - T) \cdot \left(\frac{d+1}{2}\right) \cdot AV$$

where the summation is now over the total number of exceedance NET and d is the duration of the exceedance expressed in number of averaging periods, AV . Similar to AOT, the ER is expressed in *concentration x time* units, for example in *ppm.h* or *mg.h.m³*. In Table 4 next to the occurrence of exceedances, the exceedance rate averaged over the number of exceedances, is given. Examination of this table shows some - but certainly no perfect - correlation between the number of exceedances and the severity of exceedance. The data suggests that in southern countries (Greece, Italy, Portugal) the concentrations are generally much higher during an exceedance of the information threshold than in the northern countries. The steep fall-off in the number of exceedances of levels of 180, 200 and 240 $\mu\text{g}/\text{m}^3$ in northern countries compared to the more gradual decrease in southern countries as shown in Figure 2, leads to a similar conclusion.

For each of the countries the lowest and highest 50-, 98- and 99.9-percentile values observed at individual stations are presented in Table 5. In this table also information on the maximum values is included. Note that the maximum 8-hourly concentration, as reported in Table 5 is based on a moving average and may therefore differ from the values in Table 3 which are based on fixed 8-hourly periods. The Czech Republic has not submitted percentile values for moving eight-hourly concentrations; Spain has submitted no statistical information at all. Statistics for Latvia and the Netherlands are not included in the table. In case of Latvia, the data coverage for the submitting station is below the required 75%; this low data

coverage is largely caused by missing data for the period 1 January - mid April. The information on maximum concentrations and on exceedances will therefore be reliable. In the case of the Netherlands no information on data coverage has been submitted and therefore the validity of the percentile values can not be judged. Full details on percentile values and the number of exceedances at the individual stations is presented in Tables I.3 of Annex I⁵.

Figure 2: Relative change in exceedances for the threshold values of 180, 200 and 240 $\mu\text{g}/\text{m}^3$. The number of exceedances of 180 $\mu\text{g}/\text{m}^3$ is set to unity; reference period 1 January - 31 December 1997. For Spain no information on exceedances of 200 $\mu\text{g}/\text{m}^3$ has been submitted. For all countries, the number of exceedances of 240 $\mu\text{g}/\text{m}^3$ has been extracted from the information on exceedances of the threshold of 180 $\mu\text{g}/\text{m}^3$.



As ozone concentrations show a high auto-correlation, the correlation between 1-h and 8-h percentile values is no surprise. Median values for hourly and moving 8-hourly concentrations are very similar. The 98-percentile for 8-h values is generally about 10% lower than the corresponding 1-h value although this ratio is slightly different for the various types of stations and has also a meteorological dependence. Detailed information on the percentile values and maximum concentrations observed at individual monitoring stations is given in Annex I, Table I.3.

For a large number of countries additional statistical information on NO_x , NO_2 and VOC concentrations has been received. This information is primarily used here for the classification of stations. Being precursors of ozone, information on ambient levels of NO_x and VOC is essential to

⁵ Annex I is only available in computer readable form from the ETC-AQ web-site (URL: <http://www.etc-aq.rivm.nl>). In this report a description of the tables in Annex I is provided.

evaluate the effectiveness of ozone abatement strategy. The information voluntarily submitted in the framework of the Ozone Directive is, however, not sufficient for such an evaluation. The limited time series (four years or less) and generally lack of information on the local environment of the stations, hampers an analysis of a possible trend in precursor NO_x and VOC emissions. As the reported NO₂ and O₃ concentrations are not measured simultaneously, it is not possible to improve the insight in spatial variability of ozone concentration based on mapping of oxidant (sum of NO₂ and ozone) concentrations. Oxidant concentrations are representative for a larger area as oxidant is less dependent on local condition and meteorological conditions than either ozone or NO₂ (see also Annex II). An overview of the reported NO_x, NO₂ and VOC concentrations is given in Table I.4 and I.5 of Annex I.

Table 4: Occurrence of exceedances (OoE; in days) and average exceedance rate (ER; in (µg.h)/m³ for the threshold value of 180 µg/m³ and in (mg.h)/m³ for the threshold values of 110 and 65 µg/m³); reference period 1 January - 31 December 1997. na = no data available.

	number of stations (b)	Threshold value (in µg/m ³)					
		180-1h		110-8h(a)		65-24h	
		OoE	ER	OoE	ER	OoE	ER
AT	111	0.1	2.7	30.5	2.9	119	55.4
BE	25	3.2	151.7	23.1	4.7	50.4	19.4
DE	373	1.4	52.2	25.5	3.4	64.0	24.2
DK	5	0.2	0.3	8.4	0.9	78.4	18.3
ES	36	4.2	141.9	na	na	na	na
FI	11	0.0	0.0	6.9	0.5	128	42.7
FR	151	1.2	13.1	21.9	2.8	64.6	24.4
GB	71	0.7	27.5	6.5	1.0	33.0	9.1
GR	13	10.1	710.1	32.0	5.2	64.6	22.7
IE	6	0	0.0	2.7	0.3	73.3	20.7
IT	121	12.3	863.9	33.8	5.4	68.0	51.3
LU	5	1.0	17.2	26.2	4.7	70.4	34.2
NL	39	1.2	44.1	14.5	2.4	31.5	10.2
PT	11	3.1	160.9	22.8	4.1	52.8	23.1
SE	6	0	0	9.5	0.9	166	56.0
EU-15	984	2.6	136	23.2	3.1	60.3	24.8
CH	13	3.7	130	58.5	9.3	135	67.2
CZ	48	0.2	3.4	28.2	2.9	112	46.9
LV	1	0	0	6.0	0.5	55.0	12.5
NO	14	0	0	6.7	1.1	147	97.3
PL	16	0.5	11.7	15.9	1.6	79.1	29.0
SK	8	0	0	4.8	0.3	30.3	7.6

(a) based on the eight hourly value between 12.00-20.00.

(b) note that differences in the number of stations reporting for each of the threshold levels may occur.

Table 5: Range in reported 50-, 98- and 99.9-percentile values and maximum observed values (based on hourly and moving eight-hourly average concentrations, period: 1997) observed at individual monitoring stations in reporting countries (in $\mu\text{g}/\text{m}^3$); na = range in percentile values not available; ? = range in percentile values can not be calculated as data coverage is below 75%, or information on data coverage is lacking; * = additional information submitted on a voluntary basis.

1997 range	1h-P50		1h-P98		1h-P99.9*		1h-MAX	
	min	max	min	max	min	max	min	max
AT	15	97	90	143	117	176	124	228
BE	18	56	96	154	150	203	166	277
DE	8	102	63	193	99	225	109	253
DK	44	62	90	117	116	153	124	181
ES	na	na	na	na	na	na	na	na
FI	35	74	86	112	106	140	118	179
FR	11	97	82	164	111	214	26	270
GB	10	64	68	124	102	194	74	314
GR	18	56	82	167	117	266	137	383
IE	40	66	82	106	na	na	114	173
IT	11	86	51	186	98	237	121	271
LU	20	62	79	146	115	182	131	203
NL	?	?	?	?	?	?	67	266
PT	10	65	44	157	63	177	77	271
SE	59	69	96	116	114	147	118	169
CH	14	87	116	163	142	205	150	274
CZ	21	80	82	140	106	171	127	200
LV	?	?	?	?	?	?		141
NO	52	70	88	111	102	146	108	162
PL	22	82	90	127	127	185	136	222
SK		50		113		130	122	161
1997 range	8h-P50		8h-P98		8h-P99.9*		8h-MAX	
	min	max	min	max	min	max	min	max
AT	15	97	83	141	108	167	112	174
BE	20	56	88	175	134	202	143	226
DE	14	102	55	180	82	219	93	225
DK	44	62	86	114	109	147	112	158
ES	na	na	na	na	na	na	na	na
FI	35	75	81	110	92	133	102	147
FR	14	97	75	159	101	194	15	211
GB	12	66	62	118	92	178	60	200
GR	20	57	72	147	97	203	109	252
IE	39	66	79	104	na	na	110	162
IT	48	48	129	129	162	162	94	230
LU	22	62	72	140	103	166	110	175
NL	?	?	?	?	?	?	57	199
PT	11	65	38	154	53	165	57	233
SE	59	69	96	113	112	142	114	147
CH	na	na	na	na	na	na	135	209
CZ	24	80	74	134	97	160	107	187
LV	?	?	?	?	?	?		129
NO	51	70	88	108	101	142	102	147
PL	23	81	81	124	114	157	125	175
SK		50		105		120	108	149

4. Discussion

4.1. Geographical coverage of monitoring stations

For 1997 information of 984 stations in EU Member States and of 92 stations in five⁶ other European countries was received; for nearly all of them geographical co-ordinates are available. For the interpretation of ozone data it is essential to have an indication of the immediate surroundings of the station as the ozone concentration may be strongly influenced by local conditions. For example, the ozone concentrations may be scavenged by locally emitted nitrogen oxides or by enhanced dry deposition as might be the case under a forest canopy; see Annex II for a brief discussion on ozone phenomenology. For nearly all stations information on immediate surroundings was made available.

As was observed in previous reports, not only the relative fractions of street, urban and rural stations but also the station density varies strongly between the reporting countries (De Leeuw *et al.*, 1997; Beck *et al.*, 1998), as Figure 3 shows. Here the station density as function of various indicators is given. The number of stations per capita or per km² land area varies strongly. The station density relative to the Gross Domestic Product seems to be slightly more constant. When comparing the number of stations with ozone precursor emissions (here the sum of NO_x and twice the VOC emissions is selected as indicator for ozone formation, see Sluyter, 1995), again a wide spread is observed. More detailed indicators (number of urban stations (excluding street stations) per urban dweller, number of rural stations per km² show even larger spread. It has to be stressed that a certain variation in densities is expected over Europe as national conditions with respect to meteorology, topography/orography, economic structure, physical planning etc. are different over Europe but the large variations observed here suggest that basic principles in defining a monitoring strategy differs strongly between the countries. In all cases the densities are normalised to a unit density averaged over all 20 reporting countries.

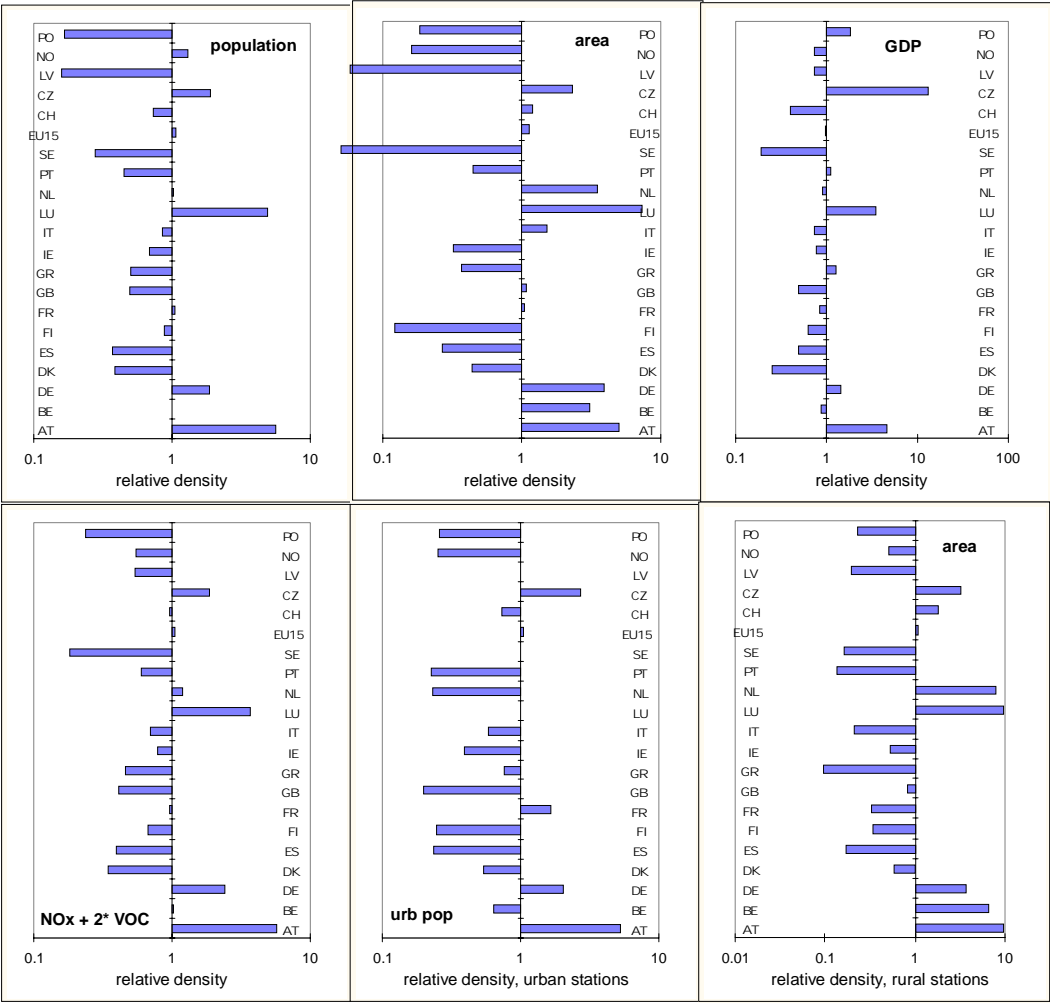
Map 2 shows the location of background monitoring stations. Tentatively, the area for which the measurements at these stations are assumed to be representative is indicated with a circle using a radius of 100 km. This 'radius of representativeness' might be different for the various regions in Europe - it might even depend on the wind direction - and should be based on more detailed analysis of the ozone phenomenology at the stations. Although the identification of the background stations is by far not complete and the radius of representativeness may differ from the assumed 100 km, Map 2 suggests that the present set of stations covers 40 to 50% of the area of the reporting countries. As is shown in Map 2 the geographical coverage of background stations is rather adequate in north-west and Central Europe but in southern regions gaps are noted. In Scandinavia the

⁶ In this chapter data from Slovakia has not been included.

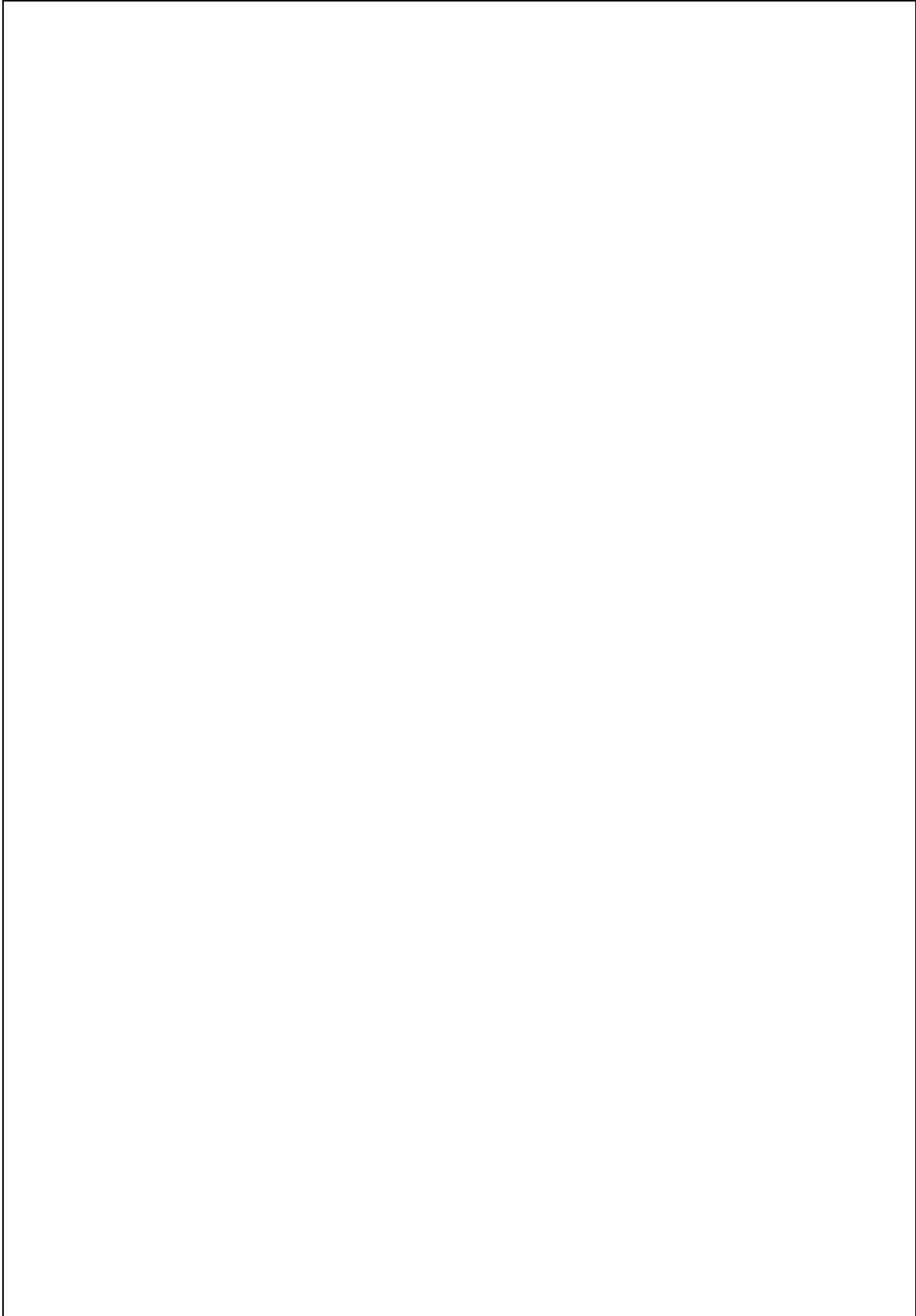
coverage by background stations seems to be inadequate but the small gradients in ozone concentrations over this region suggest a larger radius of representativeness than the assumed 100 km.

To compare the monitoring results in the various countries it is important that in the national or regional networks similar criteria are used to classify the stations. Further work on harmonisation of station classification schemes is undertaken by ETC/AQ in co-operation with DGXI (Larsen *et al.*, 1998). A second requirement for obtaining Europe wide comparable results is the harmonisation of air quality measurements. This work is conducted on behalf of the Commission by the European Reference Laboratory of Air Pollution (ERLAP) at the Joint Research Centre in Ispra. Recently the results of a comparison of calibration procedures used by national laboratories in EU and EFTA countries were published (De Saeger *et al.*, 1997). 21 national laboratories made use of the facilities of ERLAP to check and compare their ozone calibration methods with the ERLAP reference methods. During the inter-laboratory tests the most commonly used technique for automatic measurements was based on UV absorption photometry. With the exception of two analysers, the inter-comparisons showed that measurements of various ozone concentrations were in good agreement (within 3%) with the standard value. The good agreement between the calibration techniques and results obtained by the participating laboratories shows the high level to which techniques for the measurement of ambient ozone concentrations are harmonised throughout Europe. However, periodical checks and maintenance of the analysers is essential to avoid interference with other water vapour and other trace gases.

Figure 3: Station density (the average density for all countries is normalised to unity) relative to population, land area, Gross Domestic Product and precursor emissions. The second and third graph on the bottom line show the density of urban station relative to the urban population and the number of rural stations per km², respectively. Average densities for the reporting countries are respectively: 2.5 station per million inhabitants; 0.27 station per 1000 km²; 0.14 station per 10⁹ US \$ (1990 prices); 0.026 station per emitted kton; 1.26 urban station per million urban inhabitants; 0.08 rural station per 1000 km².



Map 2: Location of background ozone monitoring stations. The area for which the ozone measurements might be representative is tentatively indicated with a circle with a radius of 100 km.



4.2. Annual statistics, 1997

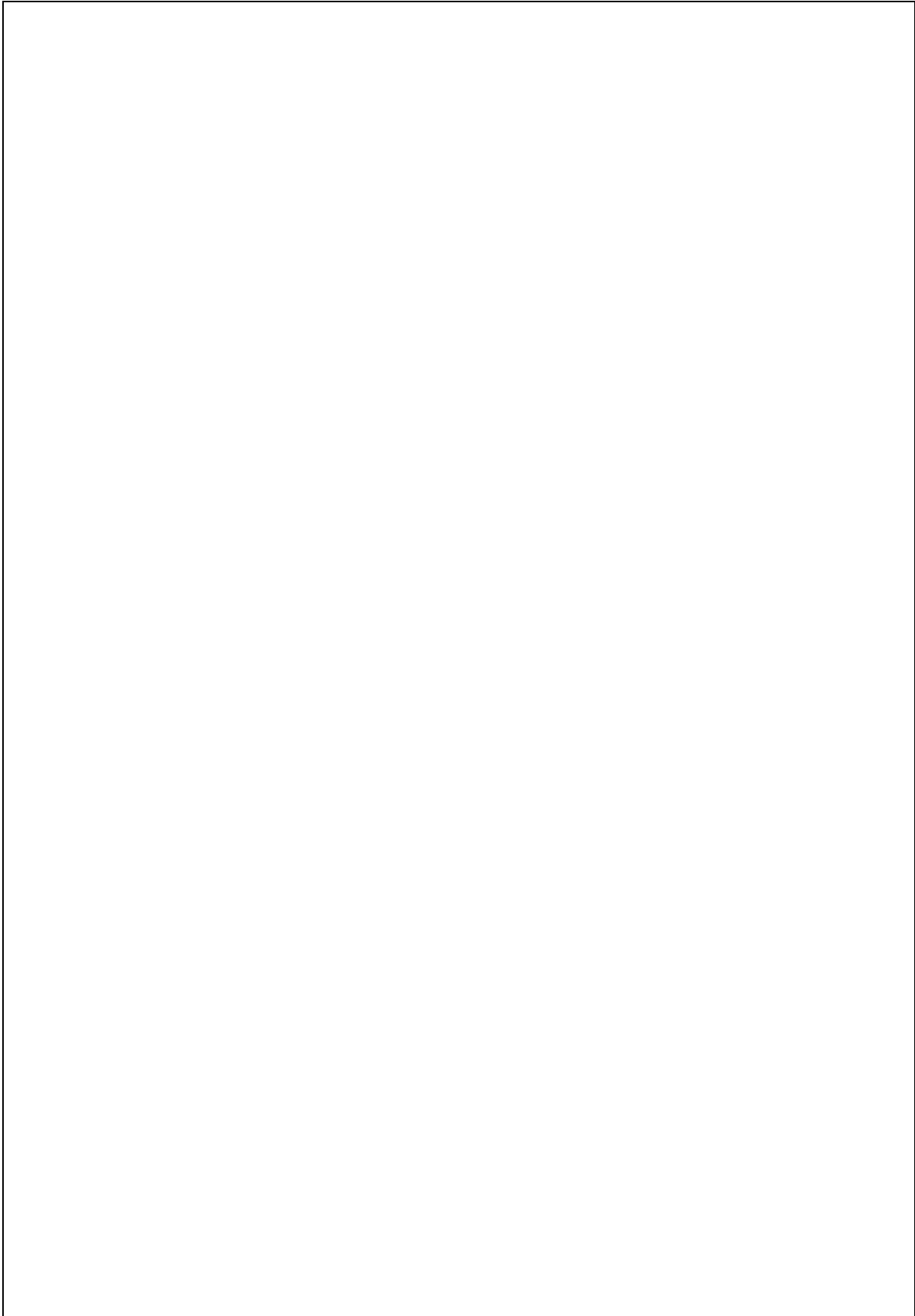
The geographical distribution of 98-percentile values calculated on the basis of hourly concentrations is presented in Map 3 for background stations and in Map 4 for urban, street and other stations.

Similar to the observations in previous years (de Leeuw *et al.*, 1997) the 98-percentiles at background stations show in general low values in the Nordic countries, and an increase from Northwest to Central Europe. In particular for the stations in Austria, the elevated location of the monitoring stations may play a role. Similar patterns have been estimated from measurements made within the framework of EMEP (Hjellbrekke, 1996). The relatively low values observed in the EMEP data over the Iberian peninsula can not be confirmed (or rejected) with the present observations. Information for Spain is not available; in Portugal there is only one rural station available for comparison. Here a 98-percentile of $157 \mu\text{g}/\text{m}^3$ is observed. This is slightly higher than the observed range of $116\text{-}149 \mu\text{g}/\text{m}^3$ at EMEP stations in Spain and Portugal in 1995; the limited comparison and the difference in measuring period precludes any conclusion.

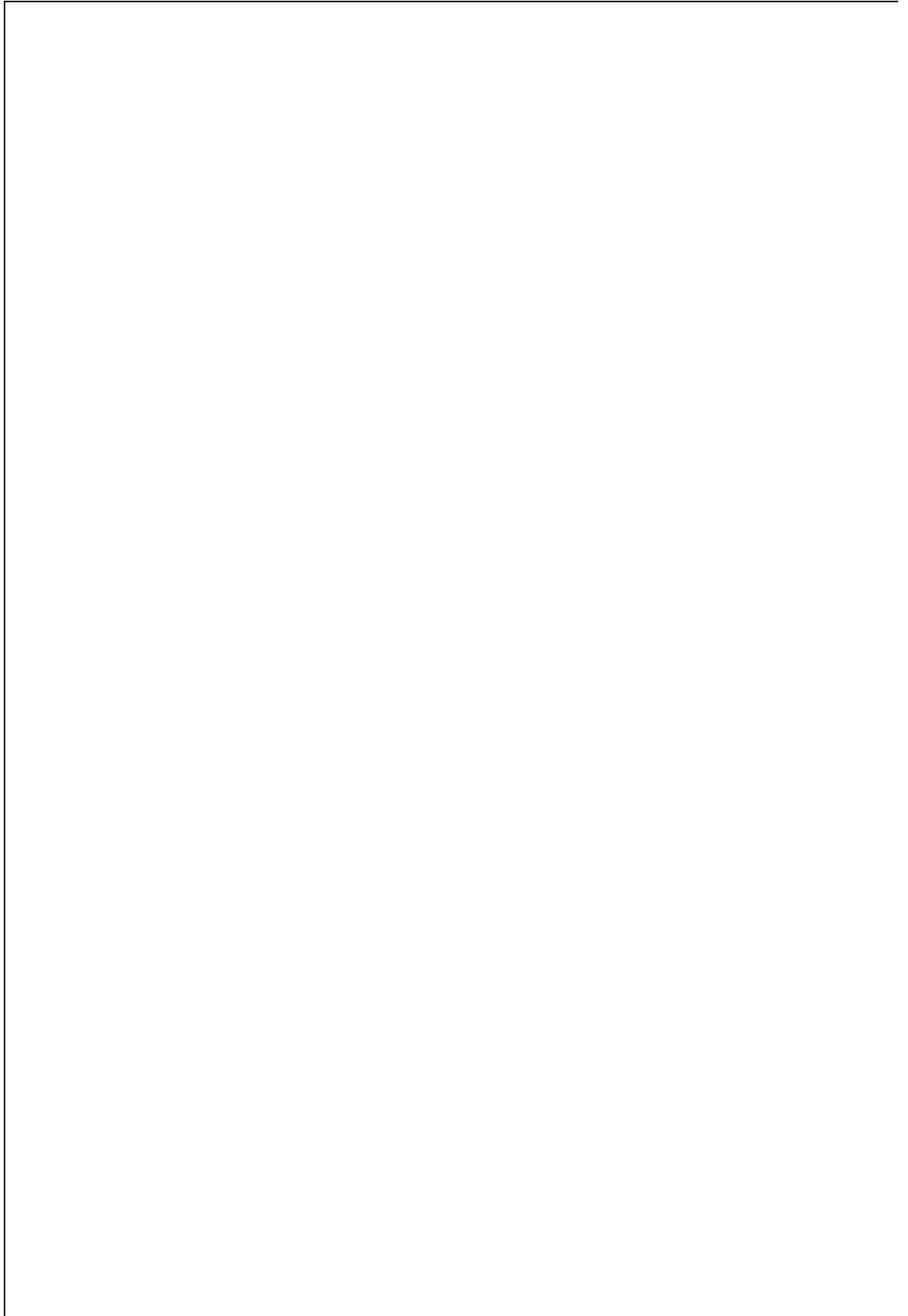
For urban and 'other' stations (Map 4) no large scale concentration gradient in 98-percentiles is recognised; high values are observed all over Europe. The local conditions seems to be more important than European-wide smog episodes (at stations downwind of the urban area relatively high ozone values might be observed whereas at stations with NO_x sources, such as traffic, in their immediate surrounding relatively low ozone levels will be measured, see Annex II).

The 98-percentile values based on moving eight-hourly average concentrations show a strong correlation with the hourly 98-percentile values: on the average, 8-h percentiles are about $7 \mu\text{g}/\text{m}^3$ lower than the corresponding 1h value. The relation between the two statistics depends on the type of the stations and also has a meteorological dependence. The geographical distribution of the 8-h percentile values is very similar to the distribution of the 1-h percentile values.

Map 3: 98 percentiles (based on hourly concentrations; $\mu\text{g}/\text{m}^3$) measured at background stations, period 1 January - 31 December 1997



Map 4: 98 percentiles (based on hourly concentrations; $\mu\text{g}/\text{m}^3$) measured at urban, street and other stations, period 1 January - 31 December 1997



4.3. Exceedances of thresholds in 1997

4.3.1. Exceedances of the threshold value for protection of human health

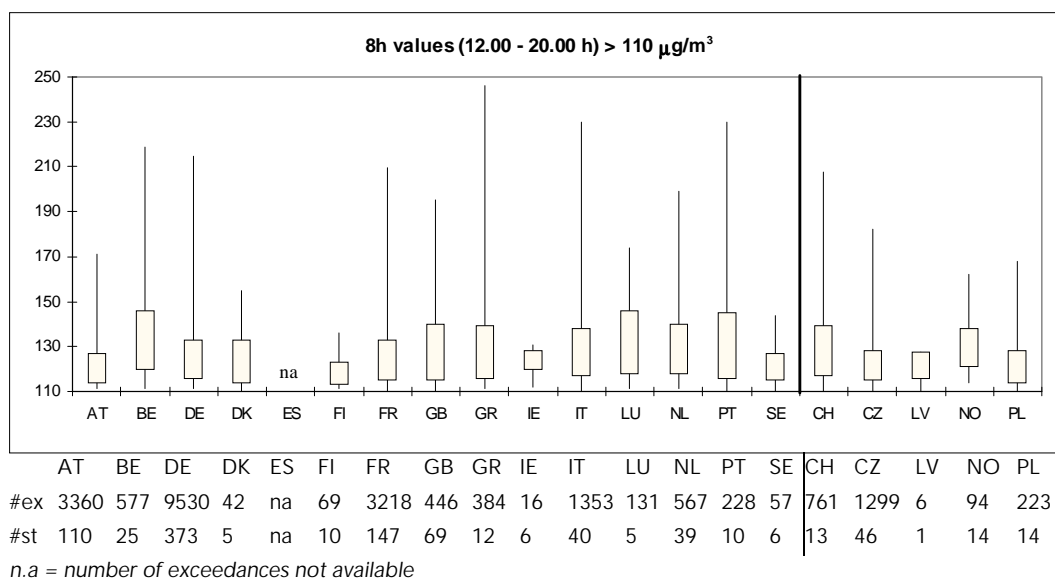
The threshold value for protection of human health ($110 \mu\text{g}/\text{m}^3$) is based on eight-hourly values. According to the Ozone Directive, four eight-hourly periods have to be considered: 0.00-8.00; 8.00-16.00, 16.00-24.00 and 12.00-20.00. Based on the average diurnal profile of ozone (see Annex II) the highest eight-hourly values are generally expected for the 12.00-20.00 period; only exceedances of the threshold values for this period have been considered here.

No clear definition of the time reference (local time or universal time, summer- or winter time) is given. This might lead to inconsistencies between the Member States. The differences between the maximum concentrations quoted for the (8h) concentration between 12.00 and 20.00 (Table 3) and for the moving (8h) average concentrations (Table 5) indicates that small errors might be introduced by the ambiguity on time reference.

In 1997 exceedances of this threshold value have been observed in all reporting countries. A maximum concentration exceeding twice the threshold value have been observed in three Member States; on a moving 8-h average base, six countries report concentrations above $220 \mu\text{g}/\text{m}^3$. The data submitted by Spain did not include information on exceedances of this threshold. The frequency by which the threshold level of $180 \mu\text{g}/\text{m}^3$ has been exceeded, suggest that, on the averaged, the level of $110 \mu\text{g}/\text{m}^3$ (8h) has been exceeded 20-30 times at a Spanish station.

Figure 4 shows the frequency distribution of eight-hourly ozone concentrations in excess of the threshold value using so-called Box-Jenkins plots. For each country the Box-Jenkins plot indicates the minimum (here the minimum is of course $110 \mu\text{g}/\text{m}^3$), the maximum, the 25 percentile and the 75 percentile value of the concentrations during exceedance. Although peaks of more than $200 \mu\text{g}/\text{m}^3$ are observed in 7 out of 19 reporting Member States, Figure 4 shows that, in more than 75% of all exceedance situations, the concentrations are below $165 \mu\text{g}/\text{m}^3$ (that is, 150% of the threshold value).

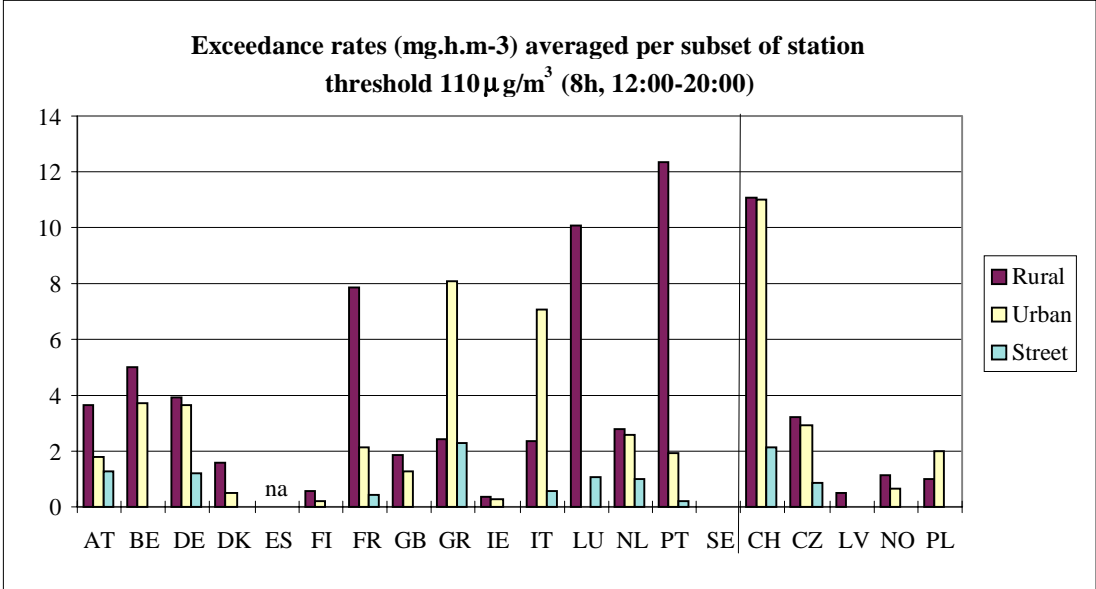
Figure 4: Frequency distribution of ozone concentrations (eight-hourly values; period 12.00-20.00; 1 January - 31 December 1997) in excess of the $110 \mu\text{g}/\text{m}^3$ threshold for hourly values. For each country the total number of observed exceedances is given in row '#Ex', the number of stations is given in row '#St'. Frequency distributions are presented as Box-Jenkins plots indicating the minimum, the 25-Percentile, the 75-percentile and the maximum value.



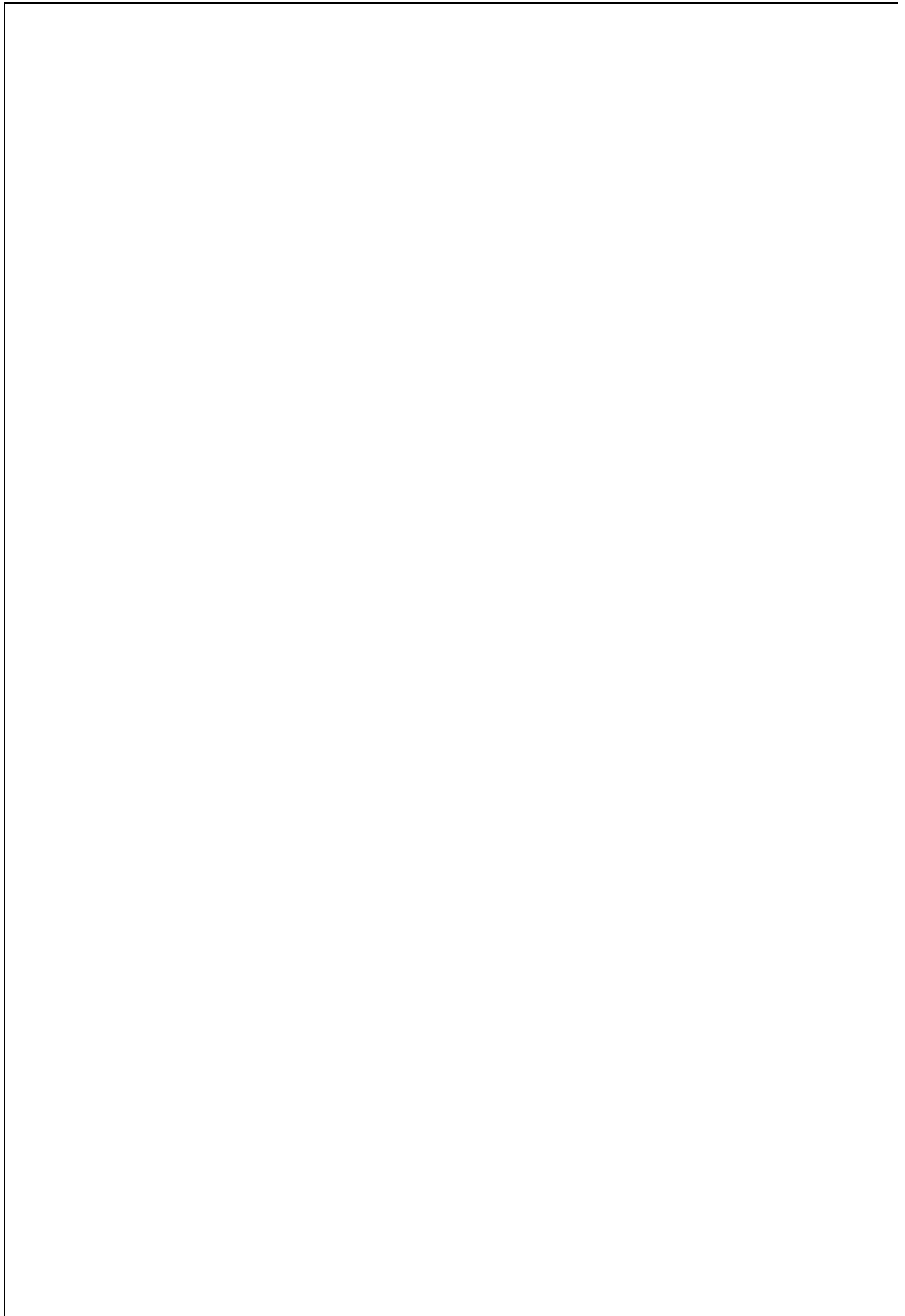
The geographical distribution of the number of days the threshold value was exceeded is shown in Map 5 for background stations and in Map 6 for urban, street and other stations. A comparison of Map 5 and Map 6 shows that exceedances are generally more frequently observed at rural stations. This is also demonstrated more clearly in Figure 5 where for each station type (rural, urban and street stations) the average exceedance rate is plotted. The average exceedance rate generally decreases in the order rural - urban - street. Exceptions are found in Greece and Italy. It might be that this is caused by a bias in the network configurations: in both countries there are much more urban than rural stations, moreover, in Italy the rural stations are all located in the most northern part on the country. Note that 'other' stations and stations for which the type has not been specified are not presented in Figure 5.

Exceedances most frequently occur in the summer months (May-August). In the winter months (January-March and September-December) exceedances have been observed very occasionally.

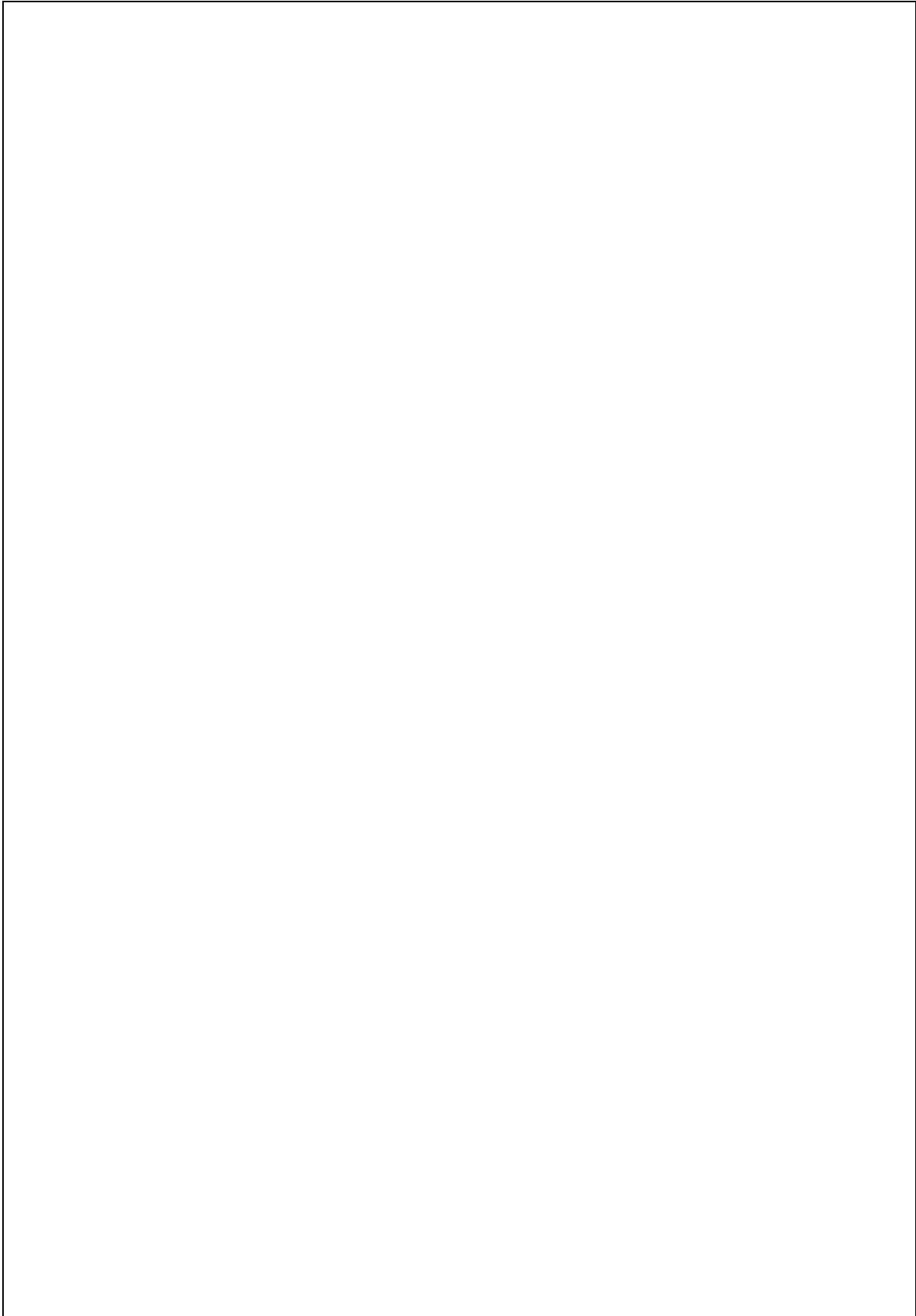
Figure 5: The exceedance rate of the threshold value for protection of human health ($110 \mu\text{g}/\text{m}^3$ for eight-hourly values (in $\text{mg}\cdot\text{h}/\text{m}^3$), period 1 January - 31 December 1997) averaged over all reporting rural, urban, and street stations. No information is available for Spain.



Map 5: Number of exceedances of the threshold value for protection of human health ($110 \mu\text{g}/\text{m}^3$ for eight hourly values) observed at background stations; 1 January - 31 December 1997; eight-hourly average values for the period 12.00-20.00.



Map 6: Number of exceedances of the threshold value for protection of human health ($110 \mu\text{g}/\text{m}^3$ for eight-hourly values) observed at urban, street and other stations; 1 January - 31 December 1997; eight-hourly average values for the period 12.00-20.00.



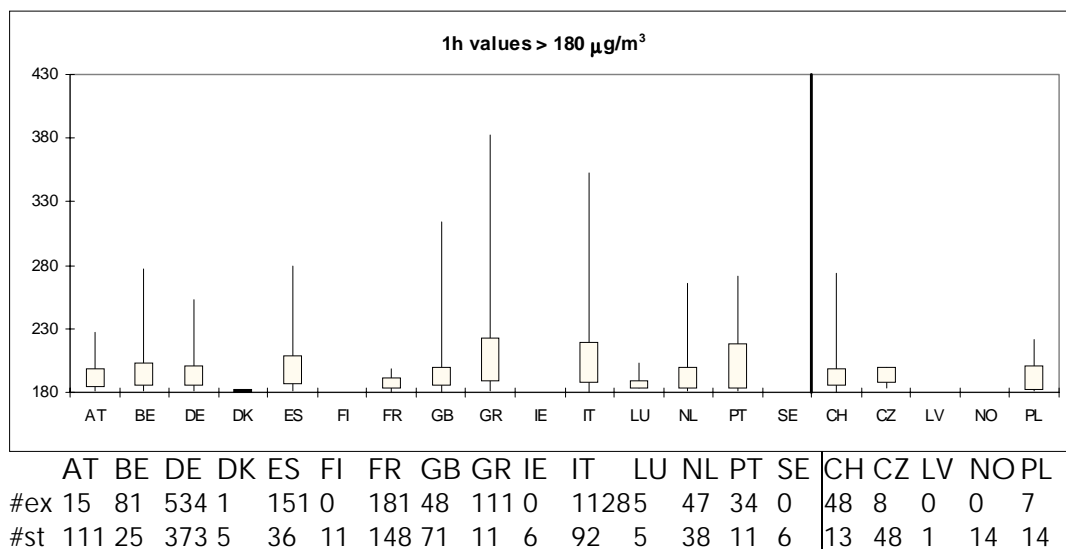
4.3.2. Exceedances of the threshold values for information and warning of the population

The threshold value for warning of the population ($360 \mu\text{g}/\text{m}^3$, hourly value) was exceeded in 1997 on 18 June, 13.00 at one station (Lykovrissi) in Athens, Greece. This day was the start of an ozone episode in Athens. The station Lykovrissi reported exceedances of the human health related threshold value of $110 \mu\text{g}/\text{m}^3$ for 8-hourly averaged during the period 18-24 June. During the whole month of June, concentrations were high in Athens: on 17 days at least one (out of seven) stations reported an hourly value exceeding $180 \mu\text{g}/\text{m}^3$. On 22 June all seven station reported an exceedance with an maximum of $229 \mu\text{g}/\text{m}^3$. For a more extensive description of this episode see de Leeuw *et al.* (1997).

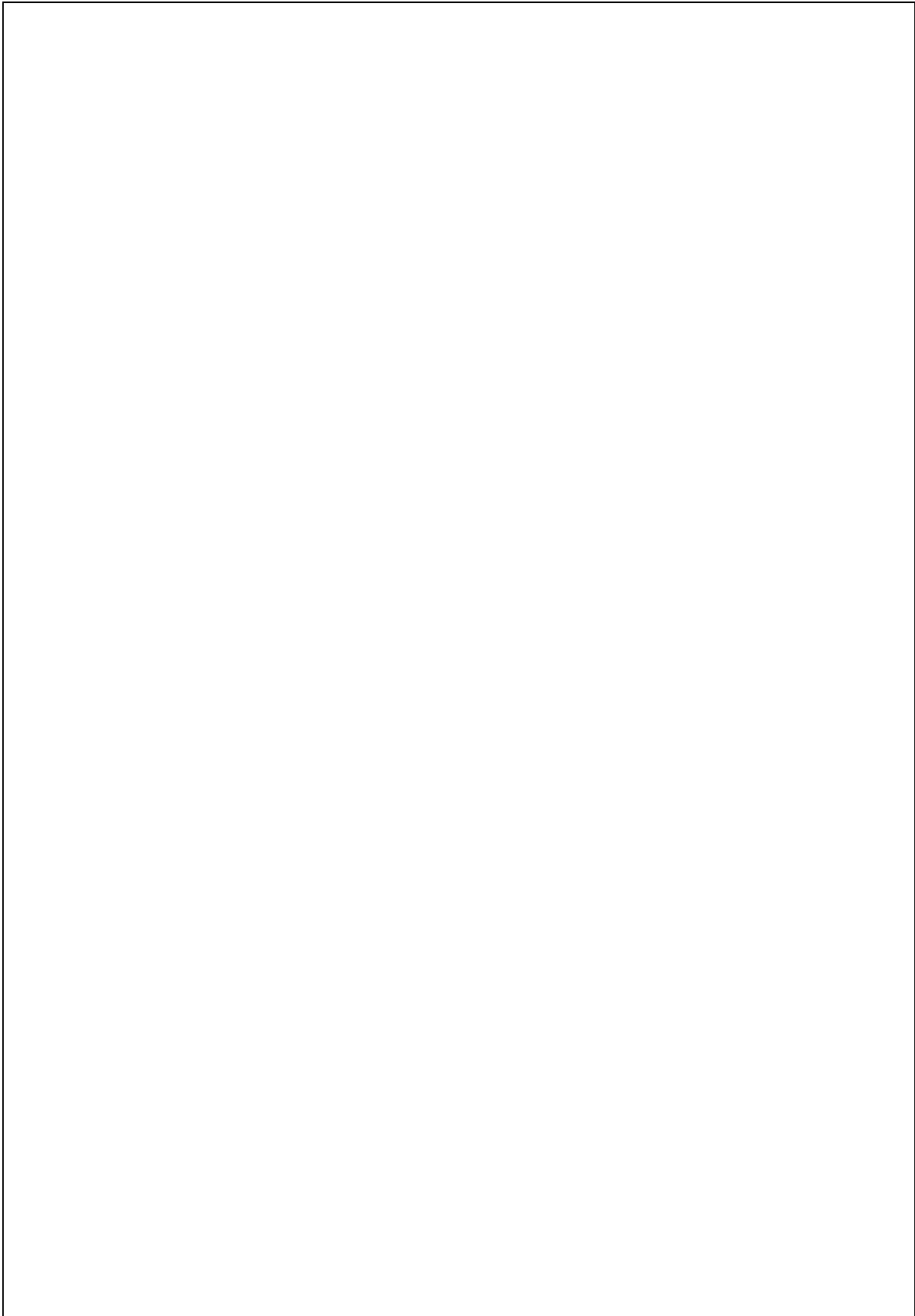
The geographical distribution of the number of exceedances of the threshold value for information of the public ($180 \mu\text{g}/\text{m}^3$, hourly value) is presented in Map 7 for background stations and in Map 8 for urban, street and other stations. Exceedances are reported by 15 countries (12 Member States). In Ireland and in the most northern countries the $180 \mu\text{g}/\text{m}^3$ level was not reached.

Figure 6 shows the frequency distribution of concentrations in excess of the threshold value. Although incidentally the threshold value may be exceeded by more than a factor of two, in almost all of the cases the exceedances are less extreme: Figure 6 shows that in nearly all Member States on 75% of the days on which the threshold value was exceeded, the level of $240 \mu\text{g}/\text{m}^3$ (that is 33% above the threshold value) was not reached.

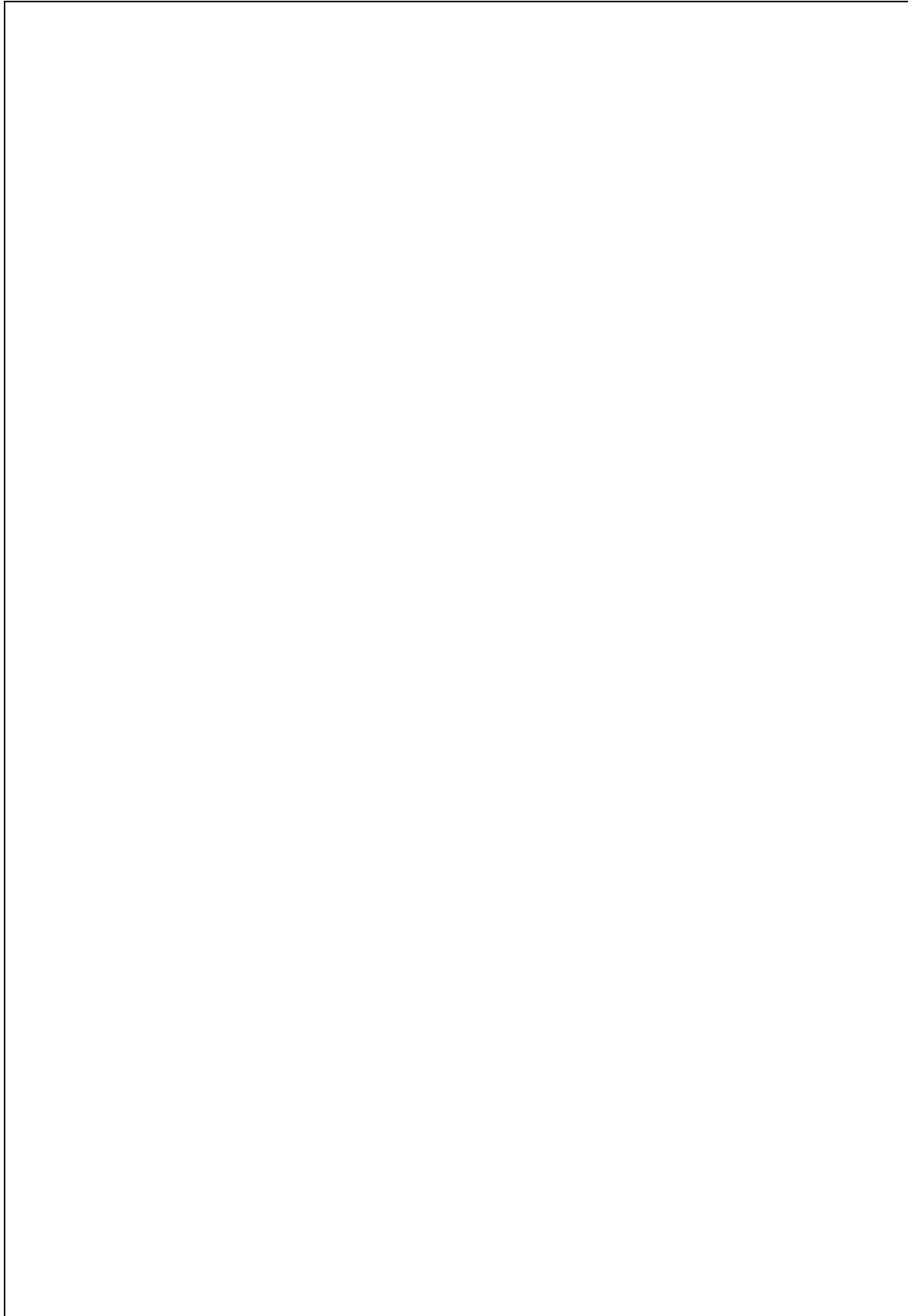
Figure 6: Frequency distribution of ozone concentrations (hourly values; 1 January - 31 December 1997) in excess of the $180 \mu\text{g}/\text{m}^3$ threshold for hourly values. For each country the total number of observed exceedances is given in row '#Ex', the number of stations is given in row '#St'. Frequency distributions are presented as Box-Jenkins plots indicating the minimum, the 25-Percentile, the 75-percentile and the maximum value.



Map 7: Number of exceedances of the threshold value for information of the population ($180 \mu\text{g}/\text{m}^3$ for hourly values) observed at background stations; 1 January - 31 December 1997



Map 8: Number of exceedances of the threshold value for information of the population ($180 \mu\text{g}/\text{m}^3$ for hourly values) observed at urban, street and other stations; 1 January - 31 December 1997



4.3.3. Exceedances of the daily threshold value for protection of vegetation

As Table 4 shows, exceedances of the daily threshold of $65 \mu\text{g}/\text{m}^3$ were frequently observed in all reporting countries. All countries have stations where the threshold was exceeded on more than 100 days in 1997 with the exception of Latvia and the Netherlands. The geographical distribution of the number of exceedances of the daily threshold is presented in Map 9 for the background stations. In Map 9 an attempt has been made to quantify the area where exceedances are observed. For the background stations a representative radius of 100 km is assumed, see also Map 2. When the 'representative areas' of two or more stations overlap, the number of exceedances in this location is estimated by a distance-weighted interpolation. At all rural stations exceedances of the $65 \mu\text{g}/\text{m}^3$ level were reported; the lowest number (5 days of exceedance) was reported for the German station Ffm. Greisheim located near Frankfurt am Main. Exceedances on more than 150 days are reported both by northern and southern Member States.

For exceedances of a daily average concentration the differences between rural and urban stations are more pronounced than is the case for hourly concentrations. In urban areas the low concentrations during the night (caused by interaction with NO_x emissions) reduce the daily average concentrations; in rural areas the decrease in ozone concentrations during the night is generally less strong. In NW Europe, however, the high NO_x emission density might cause also some quenching of ozone at rural sites which explains the relative low number of exceedances in this region compared both to North and South Europe.

Figure 7 shows the frequency distribution of daily values in excess of $65 \mu\text{g}/\text{m}^3$. Although high values of more than $200 \mu\text{g}/\text{m}^3$ have been observed, in nearly all countries for 75% of the exceedances the daily average concentration falls between 65 and $98 \mu\text{g}/\text{m}^3$ (that is 150% of the threshold value). The extremely high values reported for Italy are most likely in error; there are clear indications that at several stations the maximum observed 1h concentration was reported as daily averaged. The exceedances are most frequently observed during the growing season (May-July): more than 45% of the exceedances were reported in this three-month period. About 18% of the exceedances occurred during the winter months (January-March and October-December).

In Figure 8 the exceedance rates, averaged per subset of stations types, are given for each country. This figure clearly shows that especially at rural stations this threshold value is frequently and largely exceeded. High exceedance rates are observed both in northern and southern countries.

Figure 7: Frequency distribution of ozone concentrations (24h values; 1 January - 31 December 1996) in excess of the 65 $\mu\text{g}/\text{m}^3$ threshold for daily values. For each country the total number of observed exceedances is given in row '#Ex', the number of stations is given in row '#St'. Frequency distributions are presented as Box-Jenkins plots indicating the minimum, the 25-Percentile, the 75-percentile and the maximum value.

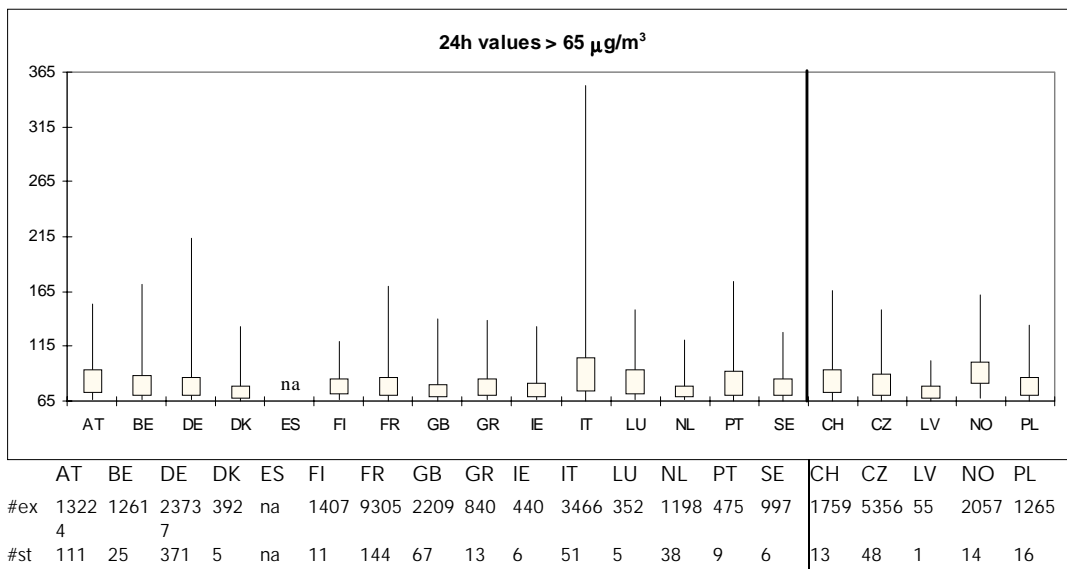
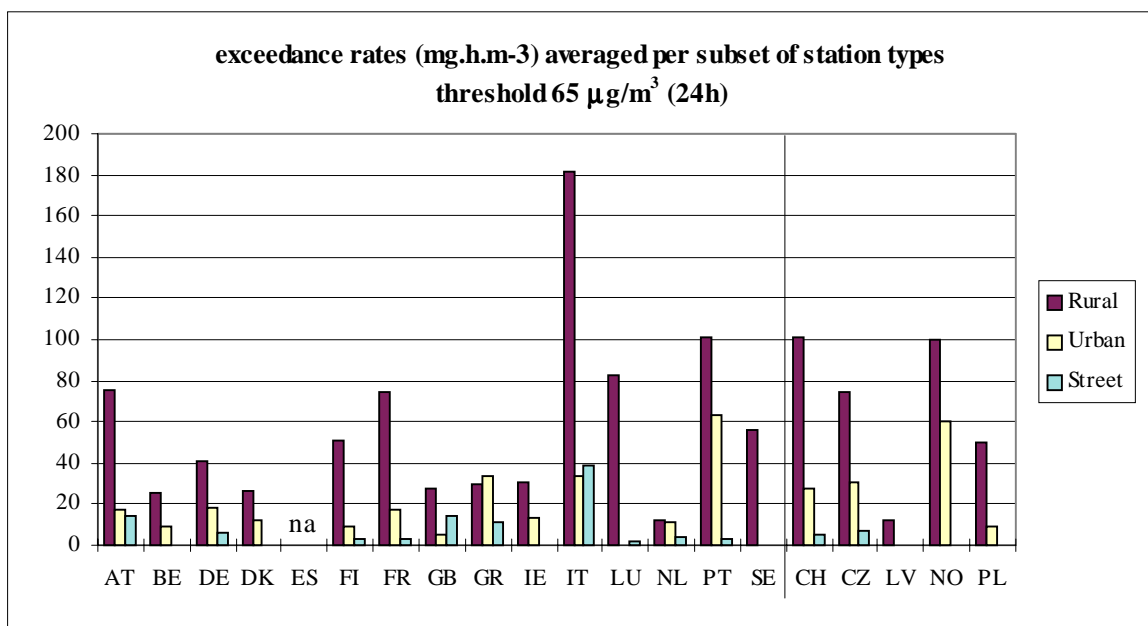
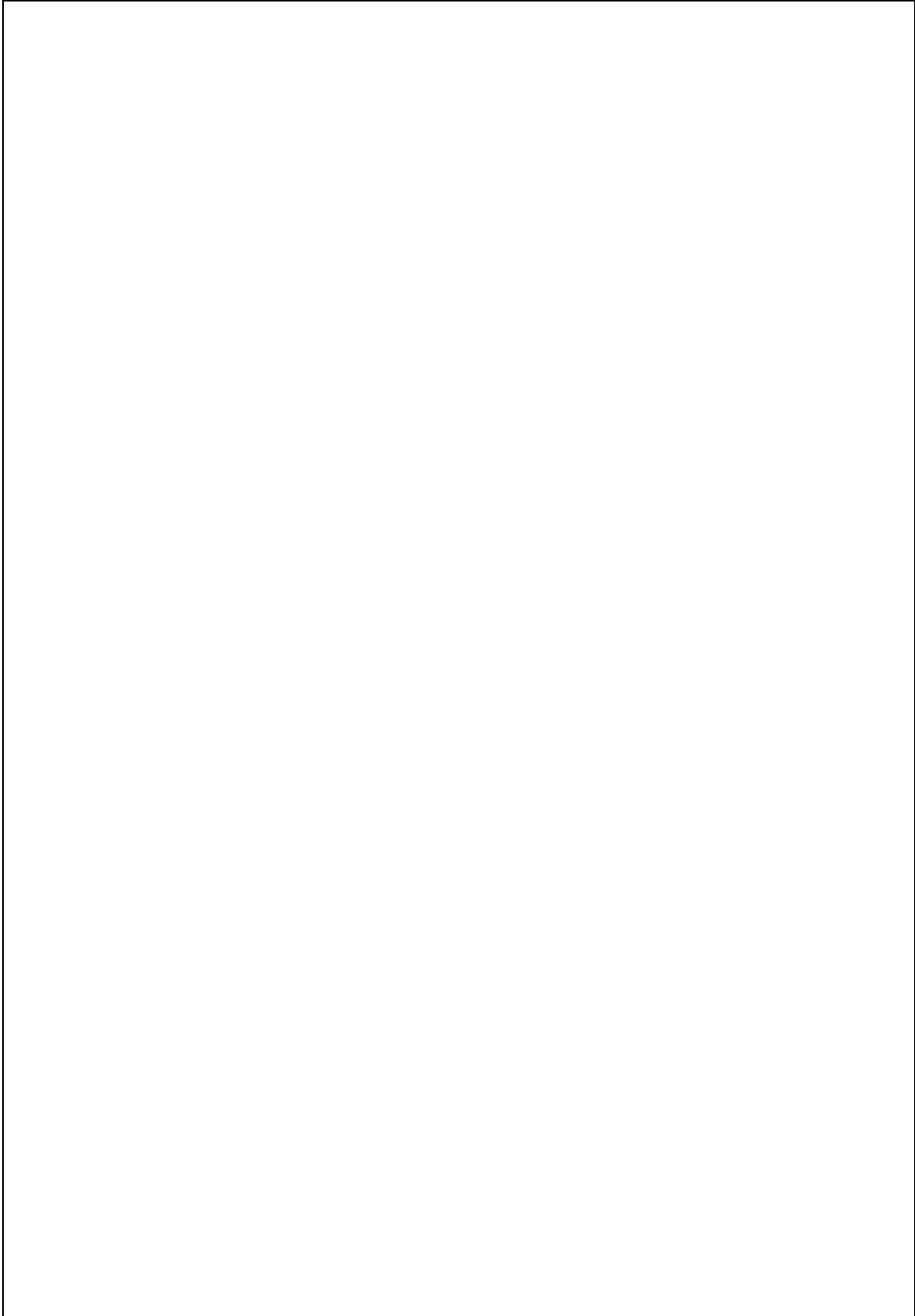


Figure 8: The exceedance rate of the threshold value for protection of vegetation of 65 $\mu\text{g}/\text{m}^3$ for daily values (in $\text{mg}\cdot\text{h}/\text{m}^3$), period 1 January - 31 December 1997) averaged over all reporting rural, urban, and street stations. No information is available for Spain.



Map 9: Number of exceedances of the threshold value for vegetation ($65 \mu\text{g}/\text{m}^3$ for daily values) observed at background stations; 1 January - 31 December 1997. Data is interpolated using inverse distance weighting and a cut-off distance of 100 km

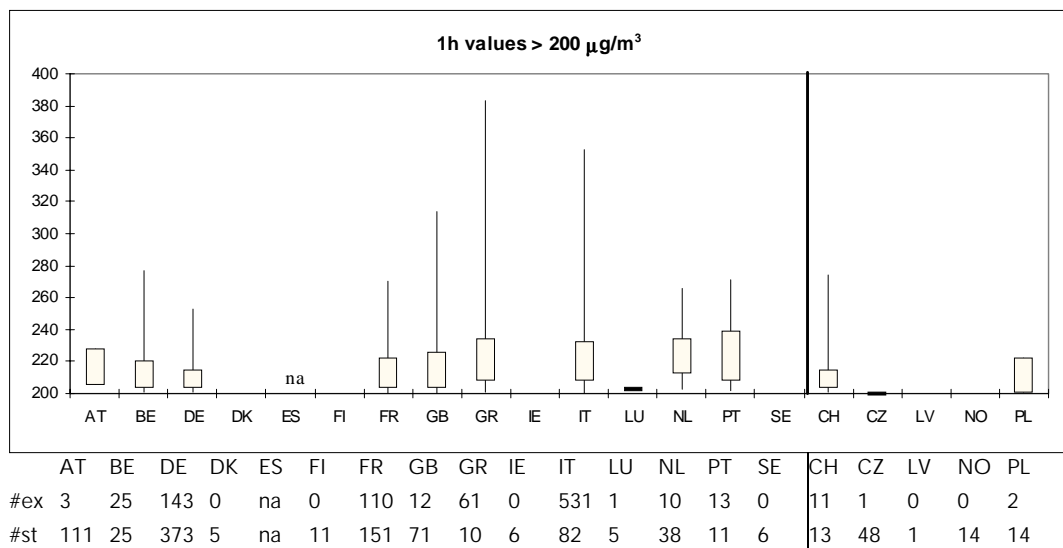


4.3.4. Exceedance of the hourly threshold value for protection of vegetation

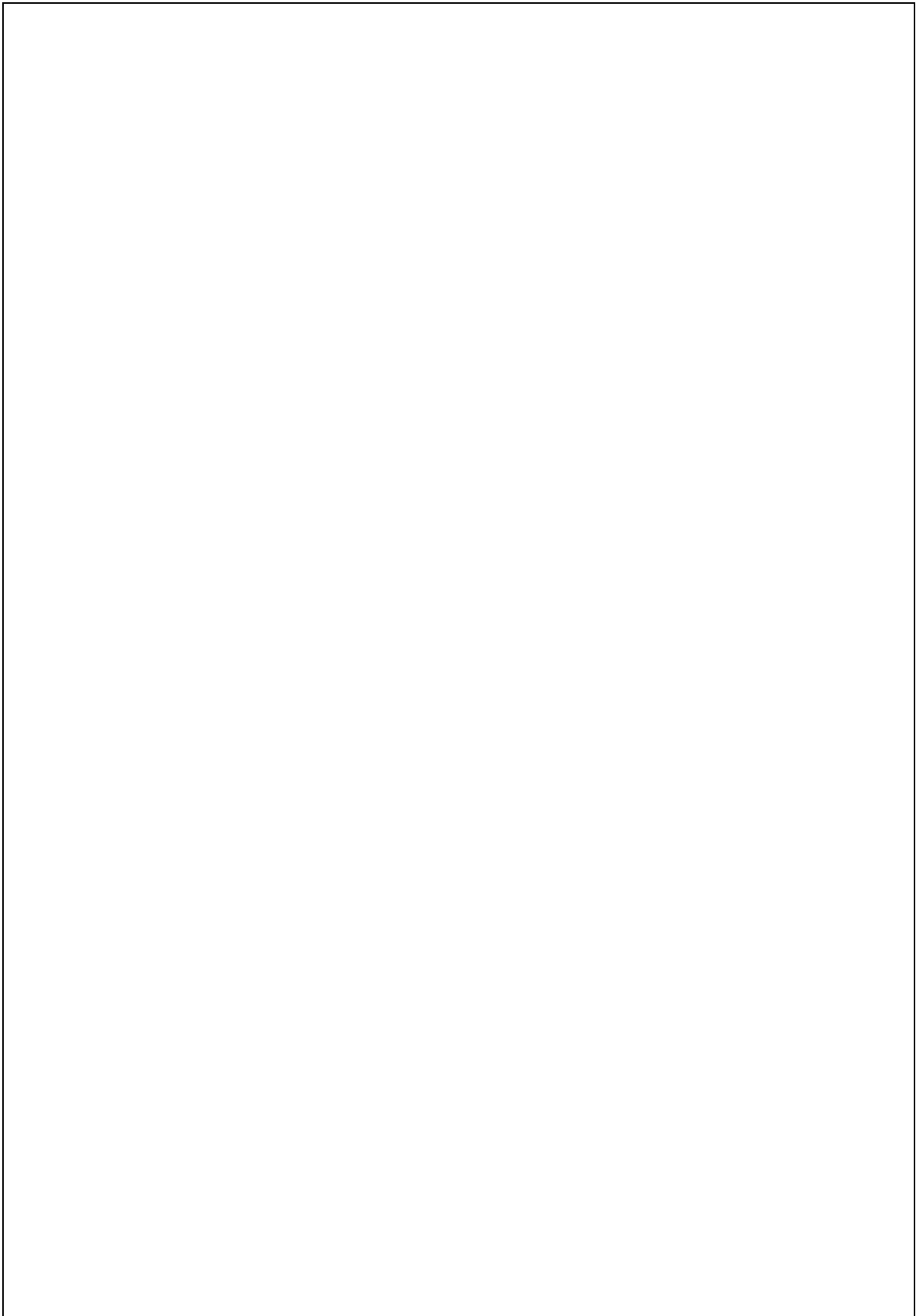
Exceedances of the hourly threshold values for protection of vegetation ($200 \mu\text{g}/\text{m}^3$) were reported by 10 Member States; no exceedances were observed in Denmark, Finland, Ireland and Sweden, see Table 4. The geographical distribution of the number of days on which this hourly threshold value has been exceeded is presented in Map 10 for background stations. The map shows that roughly above 55°N this threshold value has not been exceeded. The highest number of exceedances at a rural station (13 times) is observed at the German station Schwarzwald-Süd, see Map 10.

The frequency distributions of exceedances of the hourly threshold are presented in Figure 9. For about 75% of the exceedances the ozone levels fall between 200 and $275 \mu\text{g}/\text{m}^3$.

Figure 9: Frequency distribution of ozone concentrations (hourly values; 1 January - 31 December 1997) in excess of the $200 \mu\text{g}/\text{m}^3$ threshold for hourly values. For each country the total number of observed exceedances is given in row '#Ex', the number of stations is given in row '#St'. Frequency distributions are presented as Box-Jenkins plots indicating the minimum, the 25-Percentile, the 75-percentile and the maximum value.



Map 10: Number of exceedances of the threshold value for vegetation (200 $\mu\text{g}/\text{m}^3$ for hourly values) observed at background stations; 1 January - 31 December 1997. Data is interpolated using inverse distance weighting and a cut-off distance of 100 km



4.4. Preliminary review of exceedances of the revised guidelines of WHO

As mentioned in Chapter 1 the World Health Organization in Europe produced a revision of its guidelines for ozone (WHO, 1996a,b), see Table 6. According to the Fifth Environment Action Programme these guidelines shall serve as a basis for setting air quality standards in the Community. While these values are also relevant in the context of preparing the new daughter directive for ozone an attempt has been made to evaluate the ozone data submitted under the framework of the current directive against these guideline levels. This can only be a very preliminary evaluation as the information available under the directives deviates from the guideline requirements on averaging time and period given in Table 6.

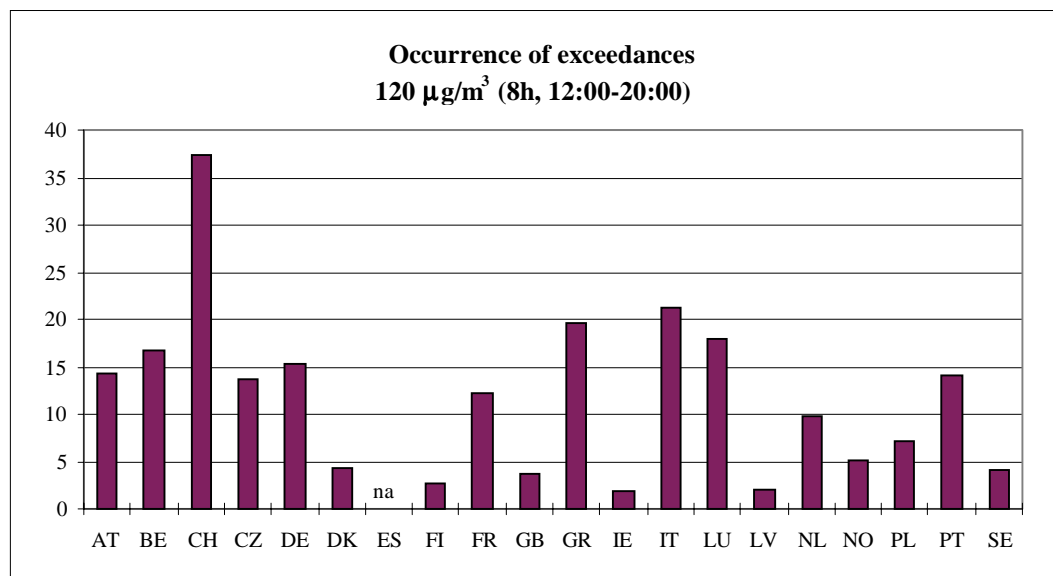
Table 6: An overview of ozone guideline levels of WHO.

Description	level	Time period	Constraints
human health	120 $\mu\text{g}/\text{m}^3$	Floating 8h mean	
for vegetation:			
Crops and semi-natural vegetation	AOT40 [*] = 3 ppmh	3 months (May-July)	
Forests	AOT40 = 10 ppmh	6 months (April-September)	
Crops (visible injury)	AOT40= 0.2 ppmh	5 days	Humid air conditions
	AOT40= 0.5 ppmh	5 days	Dry air conditions

* calculated from 1h mean values during daylight hours; 1ppmh = 1000 $\mu\text{g}/\text{m}^3\cdot\text{h}$

Referring to the guideline for human health, the exceedances reported for the 8h period between 12.00 and 20.00 has been used to evaluate exceedances of the threshold value of 120 $\mu\text{g}/\text{m}^3$ for moving 8-h values. In 1997, at one out of every five stations the 8h-averaged concentration between 12.00 and 20.00 is in excess of a level of 120 $\mu\text{g}/\text{m}^3$ on more than 20 days. As ozone concentrations generally reach their maximum in the late afternoon, the mean over the fixed period 12.00-20.00 is expected to be a good approximation of the highest daily 8-h values calculated from a moving averaged. Figure 10 shows a Europe wide exceedance of this threshold value. However, evaluation of hourly data has shown that when comparing the number of exceedance days calculated from moving 8-hourly means with such based on the fixed 12-20 h period, the latter is about 15% lower, in urban areas even 25% lower (Martin Lutz, private communication). Figure 10 might therefore give an underestimation of the exceedances of the WHO guideline value.

Figure 10: The occurrence of exceedances of the WHO guideline for protection of human health ($120 \mu\text{g}/\text{m}^3$ for moving 8-hourly values; calculations are, however, based on ozone levels reported for the fixed time period 12.00-20.00h, see text), averaged over all stations; period 1 January - 31 December 1997.



Evaluation on the AOT40 values for vegetation is not possible using the available information. A crude estimate might be based on the information on exceedance of a daily concentration of $65 \mu\text{g}/\text{m}^3$. The *exceedance rate* (see Eq 2) of a threshold of $80 \mu\text{g}/\text{m}^3$ (ER40) and the AOT40 values differ, however, on the time window for integration: the AOT40 is based on hourly values during the 'daylight'-period (8.00-20.00 CET) whereas the ER40 is based on 24h per day. A longer integration time might lead to higher values. On the other hand, days with a daily average below $80 \mu\text{g}/\text{m}^3$ do not contribute to the ER40 but may contribute to the AOT40 when hourly averages peak above $80 \mu\text{g}/\text{m}^3$. Due to these differences the exceedance rate may either over- or underestimates the AOT40. A preliminary analysis using hourly data for a limited number of stations in Latvia, the Netherlands, and the United Kingdom showed that the ER40 is lower than the corresponding AOT40; in some cases AOT40 is even a factor 3-8 higher than the ER40.

An exceedance rate of $6000 \mu\text{g}/\text{m}^3 \cdot \text{h}$ was exceeded in 1997 on 26% of all reporting stations; limiting the analysis only to the rural stations, this percentage increases to 41%. In view of the discussion above this will give a severe underestimation of the stations where the WHO critical level for protection of crops and semi-natural vegetation (AOT40 = $6000 \mu\text{g}/\text{m}^3 \cdot \text{h}$) is exceeded. Both observations as well as model calculations made within EMEP (EMEP MSC-W, 1998) show that south of $55-57^\circ\text{N}$ an AOT40 of $6000 \mu\text{g}/\text{m}^3 \cdot \text{h}$ is exceeded in more than 90% of the area.

It must be concluded that the information currently collected under the Ozone Directive is not adequate to assess exceedances of the revised WHO-guidelines for ozone.

4.5. Data reported for 1994-1997

Trends in ozone concentrations are expected to result from trends in precursor emissions in Europe and from the increasing trend in hemispheric background concentrations (Borrell and van den Hout, 1995). The magnitude and even the sign of a possible trend might differ from location to location. In a study of trends in concentrations of ozone and related species in the Netherlands and nearby countries Roemer (1996) concluded that the ground level oxidant (sum of ozone and NO₂) concentrations have decreased significantly in the Netherlands from 1981 to 1994 with an average decrease of about 1% per year. For Germany a slightly downward (in the northern part) or slightly upward (in the southern part) trend was noted but probably none of these trends is significant at the 95% confidence interval (Roemer, 1996).

From an analysis of ozone concentrations in 'unpolluted' air masses arriving over the North Atlantic region at the Irish station Mace Head, it appeared that over the 1990-1996 period there is an upward trend of 0.092 ppb per year (or 0.2% per year) in background ozone concentrations (PORG, 1997). Model calculations on a global scale support the findings of an ongoing change in chemical composition of tropospheric background air. In the late 1990s daily mean concentrations of ozone are three to four times higher than in the pre-industrial era as a result of the tremendous growth in NO_x emissions from industry and the transport sector.

The year to year variation in 50-percentile values are relatively small (2-7 µg/m³ on a P50-value of about 30 to 60 µg/m³) when compared to the variations in the 98-percentile values (a variation of 11-18 µg/m³ for P98-values ranging from 76-145 µg/m³). Peak values of ozone are strongly correlated with temperature mainly because the conditions leading to high temperatures (e.g. strong solar radiation, low wind speeds, continental flows) also trigger photochemical formation. Meteorological fluctuations may cause variations in peak ozone levels that are much larger than the variations due to changes in precursor emissions. A yearly fluctuation of about 15% in 98-percentile value is not exceptional.

The time series reported in the framework of the Ozone Directive, now covering a period of four years (1994-1997), are still too short or too variable in terms of station configuration to give conclusive answers on an ozone trend in the whole of the EU. Figure 11 shows the number of occurrence of exceedance (that is, the number of exceedances averaged per station) for the 110 µg/m³ and the 65 µg/m³ level over the four-year period. Judging the exceedances of the 110 µg/m³ threshold, 1997 is a year with generally a low number of exceedances and a decreasing trend is observed in some countries (e.g. AT, DE) but not in all (see, for example IT). A different picture is found for exceedances of the daily threshold of

65 $\mu\text{g}/\text{m}^3$. Here, 1997 is not a year with an exceptionally low number of exceedances; for a number of countries (AT, BE, FR, IT, PT, SE) 1997 is even the year with the highest numbers. Data for DK, GB, and GR suggest on the other hand, a decreasing trend.

For various Member States data reports are available for years previous to 1994. Longest time series are available for Belgium, Luxembourg, the Netherlands, and United Kingdom. At the level of the individual stations in these countries, a possible trend in ozone levels has been examined using the non-parametric Mann-Kendall test (Gilbert, 1987). Only those stations for which at least data for 5 years are available have been included in the tests. The results for trends in 50- and 98-percentile values are summarised in Table 6. For most of the stations no significant trend in 50-percentile values is observed. For the stations with a significant trend, the results are mixed. An upward trend of 1-3 $\mu\text{g}/\text{m}^3$ per year is seen at a few stations in Belgium and Luxembourg. Trends in the United Kingdom are both positive or negative. The same is true for the Netherlands, although here the number of stations with a downward trend outweighs the number of stations with a positive trend. In the analysis of the 98-percentiles also no clear picture for a country or region emerged. Only for the Netherlands the majority of stations show a downward trend but here artefacts caused by the introduction of a new ozone monitor during the winter of 1990/1991 might not have been excluded (RIVM, personal communication).

Figure 11: Occurrence of exceedances of threshold values of 110 (8h) and 65 (24h) $\mu\text{g}/\text{m}^3$ in EU Member States during the period 1994-1997.

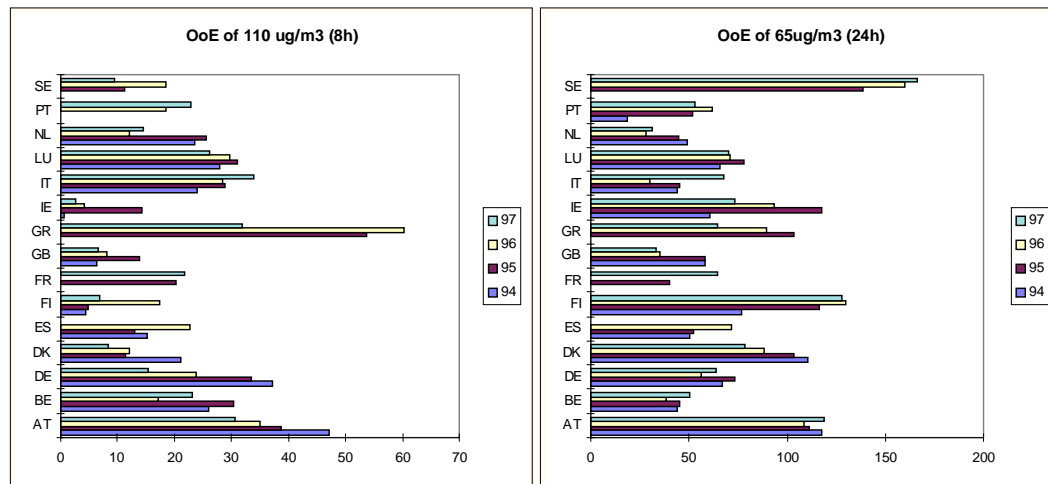


Table 6: Trend estimates for 50- and 98-percentile values for individual stations in Belgium, United Kingdom, Luxembourg and the Netherlands; period 1989-1997.

member state	number of stations ⁽¹⁾	slope range ($\mu\text{g}/\text{m}^3$ per year) ⁽²⁾
50-percentile		
BE	7(3,0)	1.6 \leftrightarrow 3.3 (0.0 \leftrightarrow 3.3)
GB	24 (2,2)	-1.0 \leftrightarrow 1.4 (-1.0 \leftrightarrow 1.4)
LU	5 (2,0)	0.9 \leftrightarrow 2.6(-1.8 \leftrightarrow 2.6)
NL	32(3,10)	-1.6 \leftrightarrow 0.9 (-1.6 \leftrightarrow 1.0)
98 percentile		
BE	7 (1,0)	1.5 (-2.5 \leftrightarrow 8.3)
GB	24 (0,2)	-6.7 \leftrightarrow -6.0 (-6.7 \leftrightarrow 6.0)
LU	5 (1,0)	1.8 (-0.3 \leftrightarrow 3.0)
NL	32 (0,20)	-6.3 \leftrightarrow -2.6 (-6.3 \leftrightarrow 3.0)

(1) in parentheses the number of stations with a significant upward, respectively downward trend (at an $\alpha = 0.10$ significant level) is given;

(2) range in estimated annual trends for stations with significant trend; results for all stations including those where no significant trend has been observed are given in parentheses.

5. Conclusions and recommendations

1. **Data for 1997 was received by the European Commission from all Member States. In addition, information for 6 other European countries was received.**

The 15 EU Member States provided information on ozone concentrations for a total of 984 monitoring stations. Switzerland, the Czech Republic, Latvia, Norway, Poland and Slovakia provided information on ozone concentrations for 100 stations.

2. **The threshold value set for the protection of human health was exceeded substantially in all reporting countries.**

The threshold value of $110 \mu\text{g}/\text{m}^3$ (8h-average) was exceeded substantially (in about 25% of the reported exceedances the 8h-average concentrations exceeded $165 \mu\text{g}/\text{m}^3$).

3. **The threshold values set for the protection of vegetation were exceeded substantially and in almost all EU Member States.**

The threshold value of $65 \mu\text{g}/\text{m}^3$ (24h average) is reported to be exceeded substantially (by up to a factor 3), widely (in all reporting countries) and frequently (nearly all countries report exceedances during more than 100 days at one or more of their stations). The threshold value of $200 \mu\text{g}/\text{m}^3$ (hourly average) is exceeded largely and widely (in total 13 countries, 10 EU Member States) on a limited number of days.

4. **The threshold value for information of the population was exceeded in almost all EU Member States during a limited number of days.**

Exceedance of the information threshold value of $180 \mu\text{g}/\text{m}^3$ (1 h average) was reported for stations in 15 countries (12 EU Member States). For one station (Lykovrissi, Athens) an exceedance of the warning level of $360 \mu\text{g}/\text{m}^3$ (1h average) was reported.

5. **The spatial coverage and quality documentation of the data need further improvement.**

Depending on the local situation, the ozone monitoring stations are characterised as rural, urban, street or other (e.g. industrial). Further work towards a harmonised classification system is recommended.

The present subsets of rural stations and urban stations are not representative for the land area of the EU and the urban population, respectively.

Spatial coverage and documentation of the monitoring data quality need improvement if the level of protection of human health and ecosystems in Europe to elevated ozone levels is to be fully assessed. Member States are encouraged to reconsider their ozone measuring

networks in the light of the spatial coverage of ozone monitoring stations. It is recommended to improve the documentation on the representativity and on the surroundings of the existing stations.

6. **It is recommended to improve the reporting of ozone precursors (NO_x, NO₂ and VOC).**

NO_x measurements should be co-located with the ozone monitoring stations as NO_x can be used as an indicator of the station representativity. Moreover, precursor concentrations will be needed for testing of compliance with VOC and NO_x emission reduction programs.

Acknowledgement

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ANNEX 1: Observed exceedances and annual statistics

Observed exceedances of ozone threshold values and annual statistics in 1997 at individual monitoring stations reporting in the framework of the Council Directive 92/72/EEC on air pollution by ozone.

Information on exceedances and concentrations of ozone and its precursors for the individual stations in EU Member States and several other European countries reporting in the framework of the Ozone Directive over the period 1 January 1997-31 December 1997 is available in computer readable form only. In the framework of the Council decision on Exchange of Information (Council Decision 97/101/EC) the Member States will report hourly ozone concentration for (a selection of) the reporting stations. These hourly data will become available from the EEA-ETC/AQ database AIRBASE.

Exceedances of the threshold value of 360 µg/m³ for hourly values has been observed in 1997 at one station only:

country	station name & city	date, time	max conc (µg/m ³)
Greece	Lykovrissi, Athens	18 June, 13.00	383

From the AIRBASE pages at the ETC/AQ Website (URL: <http://etcaq.rivm.nl>) the following tables are available:

Table I.1: Number of days, the number of periods of consecutive days on which exceedances of the threshold values of 180 µg/m³ and 200 µg/m³ for hourly values have been observed and exceedance rate⁷ at reporting monitoring stations; period 1 January - 31 December 1997.

Table I.2: Number of days, the number of periods of consecutive days on which exceedances of the threshold values of 110 µg/m³ for 8-hourly values and 65 µg/m³ for daily values have been observed and exceedance rates at reporting monitoring stations; period 1 January - 31 December 1997.

⁷ To give an indication of the severity of exceedances the exceedance rate is given. The exceedance rate is defined as:

$$ER = \sum_i^{NET} (C_{i,\max} - T) \cdot \left(\frac{d+1}{2} \right) \cdot AV$$

where the summation is over the total number of exceedance *NET* and *d* is the duration of the exceedance expressed in number of averaging period, *AV*. The ER is expressed in concentration x time units, for example in ppm.h or mg.h.m³.

Table I.3. Annual statistics (50, 98 and 99.9 percentile values and maximum values) of hourly and moving 8-hourly ozone concentrations for reporting monitoring stations; all concentrations in $\mu\text{g}/\text{m}^3$; period 1 January - 31 December 1997.

Table I.4. Annual statistics (averaged, 50-, 95- and 98- percentile values) of hourly NO_x and NO_2 concentrations for reporting monitoring stations; all concentrations as NO_2 equivalents in $\mu\text{g}/\text{m}^3$; period 1 January - 31 December 1997.

Table I.5. Annual statistics (averaged, 50-, 95- and 98- percentile values) of daily VOC concentrations for reporting monitoring stations; all concentrations in $\mu\text{g}/\text{m}^3$; period 1 January - 31 December 1997.

Notes to the tables

Table I.1

In Table I.1 it is also indicated when no exceedance of (one or more) threshold values has been observed at the station. This information has not been submitted by the Member States and is inferred from the available data. In cases where the information was insufficient for an unambiguous answer a '?' is printed; '-1' indicates missing data.

Explanations of headings of Table I.1:

<i>column</i>	<i>name</i>	<i>explanation</i>
1	codcou	country code;
2	staname	name of monitoring station;
3	incod	code number of monitoring station;
4	ville	city where station is located;
5	ER-180 (1)	exceedance rate for the $180 \mu\text{g}/\text{m}^3$ threshold values for 1h average ozone concentrations;
6	ER-200 (1)	exceedance rate for the $200 \mu\text{g}/\text{m}^3$ threshold values for 1h average ozone concentrations has been exceeded;
7	D-180 (1)	number of days on which the $180 \mu\text{g}/\text{m}^3$ threshold values for 1h average ozone concentrations has been exceeded;
8	D-200 (1)	number of days on which the $200 \mu\text{g}/\text{m}^3$ threshold values for 1h average ozone concentrations has been exceeded;
9	P-180 (1)	number of periods of consecutive days on which the $180 \mu\text{g}/\text{m}^3$ threshold values for 1h average ozone concentrations has been exceeded;
10	P-200 (1)	number of periods of consecutive days on which the $200 \mu\text{g}/\text{m}^3$ threshold values for 1h average ozone concentrations has been exceeded;
11	% 1h val	percentage of valid 1h averaged ozone values.

Table I.2:
Explanations of headings of Table I.2:

<i>column</i>	<i>name</i>	<i>explanation</i>
1	codcou	country code;
2	staname	name of monitoring station;
3	incod	code number of monitoring station;
4	ville	city where station is located;
5	ER-110 (8a)	exceedance rate for the 110 $\mu\text{g}/\text{m}^3$ threshold values for the non-overlapping 8-hourly ozone values between 0.00-8.00, 8.00-16.00 and 16.00 and 24.00;
6	ER-110 (8b)	exceedance rate for the 110 $\mu\text{g}/\text{m}^3$ threshold values for the 8-hourly ozone values between 12.00-20.00;
7	D-110 (8a)	number of days on which the 110 $\mu\text{g}/\text{m}^3$ threshold values for the non-overlapping 8-hourly ozone values between 0.00-8.00, 8.00-16.00 and 16.00 and 24.00 has been exceeded;
8	D-110 (8b)	number of days on which the 110 $\mu\text{g}/\text{m}^3$ threshold values for the 8-hourly ozone values between 12.00-20.00 has been exceeded;
9	P-110 (8a)	number of periods of consecutive days on which the 110 $\mu\text{g}/\text{m}^3$ threshold values for the non-overlapping 8-hourly ozone values between 0.00-8.00, 8.00-16.00 and 16.00 and 24.00 has been exceeded;
10	P-110 (8b)	number of periods of consecutive days on which the 110 $\mu\text{g}/\text{m}^3$ threshold values for the 8-hourly ozone values between 12.00-20.00 has been exceeded;
11	ER-65(24)	exceedance rate for the 65 $\mu\text{g}/\text{m}^3$ threshold values for the 24-hourly ozone values;
12	D-65 (24)	number of days on which the 65 $\mu\text{g}/\text{m}^3$ threshold values for the 24-hourly ozone values has been exceeded;
13	P-65 (24)	number of periods of consecutive days on which the 65 $\mu\text{g}/\text{m}^3$ threshold values for the 24-hourly ozone values has been exceeded;
14	% 8h val	percentage of valid 1h average values.

Table I.3:
Explanations of headings of Table I.3:

<i>column</i>	<i>name</i>	<i>explanation</i>
1	codcou	country code;
2	staname	name of monitoring station;
3	incod	code number of monitoring station;
4	ville	city where station is located;
5	1h P50	50 percentile of hourly ozone values ($\mu\text{g}/\text{m}^3$);
6	1h P98	98 percentile of hourly ozone values ($\mu\text{g}/\text{m}^3$);
7	1h P99.9	99.9 percentile of hourly ozone values ($\mu\text{g}/\text{m}^3$) (additional information);
8	1h MAX	maximum value of hourly ozone value ($\mu\text{g}/\text{m}^3$);
9	% 1h val	percentage of valid 1h average ozone values;
10	8h P50	50 percentile of 8-hourly ozone values ($\mu\text{g}/\text{m}^3$, calculated as a moving average);
11	8h P98	98 percentile of 8-hourly ozone values ($\mu\text{g}/\text{m}^3$);
12	8h P99.9	99.9 percentile of 8-hourly ozone values ($\mu\text{g}/\text{m}^3$, additional information);
13	8h MAX	maximum value of 8-hourly ozone value ($\mu\text{g}/\text{m}^3$);
14	% 8h val	percentage of valid 8h average ozone values ($\mu\text{g}/\text{m}^3$).

Table I.4:
Explanations of headings of Table I.4 (Note that NO_x concentrations are given as $\mu\text{g NO}_2$ equivalents per m^3 ; $1 \mu\text{g NO}_2 / \text{m}^3 = 0.52 \text{ ppb}$) :

<i>column</i>	<i>name</i>	<i>explanation</i>
1	codcou	country code;
2	staname	name of monitoring station;
3	incod	code number of monitoring station;
4	ville	city where station is located;
5	AVER_x	yearly averaged NO_x concentration ($\mu\text{g}/\text{m}^3$);
6	PARP50_x	50-percentile of hourly NO_x concentration ($\mu\text{g}/\text{m}^3$);
7	PARP90_x	90-percentile of hourly NO_x concentration ($\mu\text{g}/\text{m}^3$);
8	PARP95_x	95-percentile of hourly NO_x concentration ($\mu\text{g}/\text{m}^3$);
9	%val_x	Percentage of valid 1h average NO_x values
10	AVER_2	yearly averaged NO_2 concentration ($\mu\text{g}/\text{m}^3$);
11	PARP50_2	50-percentile of hourly NO_2 concentration ($\mu\text{g}/\text{m}^3$);
12	PARP90_2	90-percentile of hourly NO_2 concentration ($\mu\text{g}/\text{m}^3$);
13	PARP95_2	95-percentile of hourly NO_2 concentration ($\mu\text{g}/\text{m}^3$);
14	% val_2	percentage of valid 1h average NO_2 values.

Table I.5:
 Explanations of headings of Table I.5:

<i>column</i>	<i>name</i>	<i>explanation</i>
1	codcou	country code;
2	staname	name of monitoring station;
3	incod	code number of monitoring station;
4	ville	city where station is located;
5	AVER	yearly averaged VOC concentration ($\mu\text{g}/\text{m}^3$);
6	PARP50	50-percentile of hourly VOC concentration ($\mu\text{g}/\text{m}^3$);
7	PARP90	90-percentile of hourly VOC concentration ($\mu\text{g}/\text{m}^3$);
8	PARP95	95-percentile of hourly VOC concentration ($\mu\text{g}/\text{m}^3$);
9	% val	percentage of valid 24h average VOC values.

ANNEX II. Phenomenology of ozone concentrations

For a better understanding of the report, some of the main characteristics of ambient ozone are summarized here. For more advanced information on ozone, its photochemical formation and its precursors the reader is referred to documents provided by DGXI (Borrell and van den Hout, 1995; Derwent and van den Hout, 1995; Beck *et al.*, 1998) and to reports prepared in the framework of UN-ECE Convention on long-range transport of air pollution (Malik *et al.*, 1996; Hjellbrekke, 1996; Barrett and Berge, 1996) and EUROTRAC (see e.g. Borrel *et al.*, 1997; Hov, 1997).

Ozone is a secondary air pollutant formed in the atmosphere under the influence of sunlight. Ozone formation occurs at all levels in the atmosphere, from ground level up to the stratosphere. Here the discussion is limited to ozone at ground level. It has been shown that under the present conditions in Europe the ozone exposure of humans, vegetation and materials leads to adverse effects.

The precursors of the ozone formation are Volatile Organic Compounds (VOC), carbon monoxide (CO) and nitrogen oxides (NO_x). VOC and CO act as 'fuels' as they are oxidized in the process; the nitrogen oxides play an important role as 'catalysts': they are not 'consumed' in the formation process but are essential for the continuation of the process. However, nitrogen oxides are consumed in side reactions by which they are further oxidized to nitric acid or nitrates. For the continuation of the photochemical oxidation process a continuous injection of nitrogen oxides is therefore necessary.

The ozone formation takes place on various time and spatial scales: on the local scale as in urban areas as Athens or Milan, on the regional scale as is demonstrated by the photochemical episodes in Central and Northwest Europe and on the hemispheric/global scale. Highly reactive VOCs are important precursors on the local and regional scale whereas the less reactive, relatively long-lived VOCs such as methane contribute to ozone formation on the global/hemispheric scale.

The role of nitrogen oxides is complex and may be different at various distances from the source. In heavily populated areas the ozone concentrations may be lower than the regional concentrations due to chemical scavenging by local nitrogen oxide emissions. This scavenging is presented by the chemical reaction:



Nitrogen dioxide (NO₂) formed in this reaction can be seen as 'potential ozone' as in the photolysis of NO₂, nitrogen monoxide, NO, and ozone are produced:



As both the time scales of the NO-scavenging reaction (R1) and of the NO₂ photolysis are relatively short, an equilibrium between the three components will be established:



Note that the sum of O₃ and NO₂ (frequently indicated as *oxidant* or *Ox*) is independent of the equilibrium. Knowledge on simultaneously measured NO_x concentrations in general and NO₂ concentration in particular is therefore important for interpretation of ozone levels. The oxidant levels are spatially less variable than the ozone levels. A mapping procedure based on oxidant levels is therefore preferred.

The time scales of photochemical ozone formation are generally longer than the time scale associated with the above reaction R1 and R2 and close to NO_x sources a decrease in ozone concentrations may be observed; at larger distances to the source the ozone levels will increase again. In Figure II.1 the ozone concentration in an air parcel passing a NO_x source is schematically presented.

One of the consequences of this interaction with NO_x is that the representativeness of ozone monitoring stations depends strongly to what extent the station is influenced by local NO_x sources: concentrations measured at a station in a traffic situation will be representative for its direct surroundings only (e.g. less than a few 100 m) whereas a background station may measure concentrations representative for an area of several tens of kilometres. The information requested for in Article 4 of the Directive should form the basis on which a representative area for each of the monitoring stations could be defined.

At ground level ozone concentrations generally show a strong diurnal variation. At night concentrations are low, both caused by removal by dry deposition and by titration by NO-emissions according to reaction R1. In the morning concentrations increase, caused not only by the sun-light induced photochemical formation but also by the downward mixing of higher, ozone-rich layers. In the afternoon both processes will become less important and concentrations will decrease again when the loss processes dominate. The maximum concentration is frequently found in the late afternoon, around 16.00. For the eight hour periods reported in the framework of the Ozone Directive, the highest value will generally be observed between 12.00 and 20.00.

Figure II.1: Schematic presentation of ozone concentrations downwind of a large NO_x source where ozone scavenging will take place.

