Ozone Position Paper

Final version

Prepared by the Ad-Hoc Working Group on
Ozone Directive and Reduction Strategy Development

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EXECUTIVE SUMMARY

BACKGROUND

This position paper is a background document to help the Commission prepare its proposal for the new directive on ozone in ambient air (one of the 'daughter directives' required by the Council Directive on the Assessment and Management of Ambient Air Quality - the 'Framework Directive'). The new ozone directive will replace the current Council Directive 92/72/EEC on air pollution by ozone.

The proposal for the new directive will form part of an integrated package proposing the EU strategy on acidification, eutrophication and ozone. The proposed ozone air quality objectives are closely linked to the National Emission Ceilings Directive, which also belongs to this package.

The paper reflects the results of discussions in the "Ad Hoc Working Group on Ozone Directive and Reduction Strategy Development", set up by the Commission in February 1997. Member States, Industry, NGOs, the World Health Organisation (WHO), the European Environment Agency (EEA) and the UN-ECE Convention on Long-range Transboundary Air Pollution (CLRTAP) were asked to appoint experts to the Group. In drafting this Position Paper, the Group assisted the Commission in its task of preparing this new legislation on ambient air quality and devising a strategy to control ozone precursor emissions. The Ad Hoc Working Group was split into two special working groups to address the assessment of risks caused by ozone and assessment methods for ozone-related air quality respectively.

As with the Position Papers on the other pollutants, final discussions on the draft of this paper were held in the Air Quality Steering Group, which encompasses an even wider spectrum of stakeholders.

1. INTRODUCTION

In the lower layers of the atmosphere, ozone is primarily formed by a complicated series of chemical reactions initiated by sunlight. These reactions, in which nitrogen oxides (NO_x, where $NO_x = NO + NO_2$) and volatile organic compounds (VOCs[1]) react to form ozone, may take anything from hours to days depending on the VOCs, and once ozone has been produced it may persist for several days. In consequence, ozone measured at a particular location may have arisen from VOC and NO_x emissions many hundreds or even thousands of kilometres away. Close to sources, nitric oxide (NO) from emissions may react with ozone to form nitrogen dioxide (NO_2), reducing ozone concentrations locally. Maximum concentrations therefore generally occur downwind of the source areas of the precursor pollutant emissions. In some regions, however, re-circulation of air masses may cause the air to reside in the region for a number of days.

Photochemical episodes of high ozone concentrations are superimposed on a baseline concentration which varies slightly throughout the year but annually averages around 60-80 $\mu g/m^3$ over much of Europe. This is made up partly of ozone transported from the stratosphere, and partly of ozone produced in the troposphere from naturally occurring and man-made precursors (in broadly equal proportions). The anthropogenic part includes contributions from sources outside Europe, in particular the USA. There is evidence that the baseline has roughly doubled since the turn of the century.

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[1] Does not encompass methane.

VOCs derive mainly from road traffic and the use of products containing organic solvents. NO_x is mostly produced by transport and combustion processes. These major sources are already subject to legislative controls that will come into effect before 2010, which means the relative importance of different emission sources will change in the future. Although NO_x and VOCs are the most important precursors of elevated levels of ozone, production of ozone can also be stimulated by carbon monoxide, methane, or other VOCs arising from plants, trees and other natural sources.

The current ozone directive 92/72/EEC, implemented in 1994, set ozone thresholds and required Member States to monitor and report exceedances of those thresholds. In recent years both the health-related threshold and the vegetation-related thresholds were exceeded substantially and in all Member States. The threshold for providing information to the public was exceeded in almost all Member States, and the warning threshold was reached occasionally.

2. BASIC LEGISLATIVE CONCEPTS FOR AN OZONE DIRECTIVE

Ozone standards in the new daughter directive will be set in accordance with the regulatory scope of the Air Quality Framework Directive (FWD). An additional principle to be incorporated is the ultimate goal set in the 5th Environmental Action Programme of avoiding throughout the Community exceedances of (WHO) critical ozone levels, i.e. levels at which harm to health and environment does not occur or is likely to be small. .

The Ad Hoc Working Group did not propose a "limit value" for ozone, as the FWD allows a "target value" to be set instead (though only for ozone). Being less strict than a limit value in terms of compliance obligation, a target value makes greater allowance for the large-scale transboundary nature of ozone and its variation with meteorology. Member States will have to take measures to attain this target value as far as possible within a given period.

However, even if technical measures were implemented to the fullest possible extent, it seems highly unlikely that critical levels would be achieved everywhere in the Community within a foreseeable period. It is therefore proposed that target values be set for human health and for vegetation as interim objectives, associated with a specified attainment period (2010). As a complementary element, a "long-term objective" set on the basis of WHO's critical levels for ozone would reflect the goal of the 5th Environmental Action Programme. Since non-attainment of the target value is not regulated in detail in the Framework Directive, it ought to be defined more explicitly in the daughter legislation. Several FWD provisions corresponding to non-attainment of the limit values could serve as an appropriate basis.

Under Article 4(2) of the Framework Directive the Commission is responsible for re-examining the scientific basis for the air quality limit values and alert thresholds. It is proposed to require that this review be undertaken and followed by any proposal for revision of any of the ozone air quality thresholds that the Commission deems appropriate within five years. Reviews should also consider the feasibility of setting a target year for attainment of the long-term objective. Given the strong link between revising the target value and further emission reductions, a review of control strategies is also deemed necessary. In updating the ozone strategy one should also take into account future requirements of abating acidification and improving air quality, especially particulate concentrations.

For the purpose of providing health advice to the public in the event of high ozone concentrations, the two-level concept of the current ozone Directive 92/72/EEC should be retained. While the FWD defines only one "alert threshold", the Directive differentiates between the whole population and groups of people who react sensitively to ozone exposure. To be as consistent as possible, the Working Group proposed establishing a lower "information threshold", defined as an alert threshold for the sensitive population, and a higher "general alert threshold", which triggers release of information for the general public.

3. RISK ASSESSMENT

Assessment of risk from ozone is based on WHO reviews and guidelines taking into account also additional results of recently published research. Since detectable health responses occur at or close to the upper bounds of ozone background concentrations, WHO did not use an uncertainty factor in determining guidelines. It set the health-related guideline value for ozone in ambient air at 120 $\mu g/m^3$ for a period of 8 hours as a level at which acute effects on public health are likely to be small. Evidence of chronic effects and mortality was considered inadequate for defining a quantitative long-term guideline. For the protection of crops and semi-natural vegetation, WHO set the guideline at a maximum AOT40[2] value of 6000 $\mu g/m^3$.hours to be calculated only over a three-month growing season and over daylight hours. A guideline for forest protection and two critical levels for visible damage to crops were also defined.

All WHO guidelines are substantially exceeded in most parts of the EU.

In some EU countries ozone standards or other thresholds are in force.

It is proposed that both the long-term objectives and the target values for ozone be expressed in terms of the WHO guidelines. For health, the proposed long-term objective is equal to the WHO guideline; for the long-term objective for vegetation protection, the guideline for crops and semi-natural vegetation was taken, albeit somewhat adapted for operational reasons:

- ***Health-related long-term objective: the maximum 8-hour average concentration is 120 $\mu g/m^3$;***

- ***Vegetation-related long-term objective: AOT40 = 3 ppm.hours (6000 $\mu g/m^3$.hours) during May-July in the periods 8-20 h (European Central Time).***

For the target values it is proposed that a certain number of days in exceedance of the long-term objective for health be allowed. The Working Group also discussed an alternative approach of setting a higher value (160 $\mu g/m^3$) with only a few exceedances. Since evidence exists for a significant health risk below this higher level, and for reasons of transparency with respect to the long-term objective (and the WHO guideline), an overwhelming majority of the Working Group preferred to set the target value at 120 $\mu g/m^3$ with a certain number of days exceeded; and for vegetation protection, to set the maximum AOT40 value for crops and semi-natural vegetation at a higher level than the long-term objective. The numerical values of the target values are given below.

As with the existing Directive, it is proposed that two thresholds be set for public information: 180 $\mu g/m^3$ (1-hour average) for the information threshold (sensitive population); 240 $\mu g/m^3$ (1-hour average) for the general alert threshold (general population).

4. ASSESSMENT OF CONCENTRATIONS

Assessment of ozone levels can begin through the network set up under the existing ozone Directive. However, this network has been found to provide insufficient cover of the EU, and substantial improvements are necessary.

The purposes of assessment are:

- Compliance checking;

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[2] AOT40 means the sum of the differences between hourly concentrations greater than 80 $\mu g/m^3$ (= 40 parts per billion) and 80 $\mu g/m^3$ over a given period.

- Air quality management and understanding the ozone pollution issue;

- Public information.

Ozone should be monitored where areas of exceedance are expected and where the relevant receptors (population and vegetation) are present. For ozone measurements the following station types are distinguished: urban, suburban, rural, rural background, and specific (special situations). For better insight into the representativeness of ozone stations, it is proposed that NO_2 be measured at at least 50% of the ozone stations.

Table A shows the proposed minimum number of stations for agglomerations and for zones where levels exceed the long-term objective over a five-year period. If the information on ozone levels is substantially improved by modelling or other methods, the density of ozone measurements may be reduced by up to one-third. Lower network densities are defined for zones that are in compliance with the long-term objective.

Table A. Minimum number of ozone stations for agglomerations and for zones where levels are above the Long-Term Objective, where measurement is the sole source of information

	Agglomerations		**Other zones**		
Population (x1000)	Urban	Suburban	Suburban	Rural	Rural background
< 250	0	0	0	1	1 station / 50 000* km^2
< 500	0	1	1	1	as an average
< 1 000	0	2	1	2	density over all zones
< 1 500	1	2	1	3	per country
< 2 000	1	3	1	4	
< 2 750	2	3	1	5	
< 3 750	2	4	1	7	
> 3 750	2	1 add. Station per 2 m. inh.	1	1 add. station per 0.5 m. inh.	

* 1 station per 25 000 km^2 for complex terrain in regions below 55ºN latitude

The reference measurement methods of the current ozone Directive can be maintained:

- analysis method: UV absorption method (ISO CD 13964);

- calibration method: Reference UV photometer (ISO CD 13964, VDI 2468, p6).

Models and other mathematical techniques are important tools for obtaining information on the spatial distribution of ozone and for monitoring the effectiveness of emission reduction measures. In addition, short-term forecast models are very useful during episodes for forecasting how ozone levels will develop.

It is recommended that NO_x and VOC concentrations be measured, too, in order to follow precursor trends and improve our understanding of ozone formation; meteorological measurements can also be very important. However, the strategy for these measurements differs from the one for ozone/NO_2 and should not described in detail in the directive.

5. EU-WIDE ABATEMENT STRATEGY AND COST IMPLICATIONS

Existing and planned measures will bring precursor emissions down by 48% for NO_x and 49% for VOCs by 2010 compared with 1990. Average population exposure to ozone is predicted to fall by around 60% as well, but this will not solve the ozone problem in large parts of Europe. The ozone levels will remain relatively high in densely populated areas of Europe where the WHO guidelines for health protection are estimated to be exceeded on 30 days and more; the same is true for the critical level for vegetation over even larger areas. Ozone is primarily a large-scale pollutant, and so the ozone problem is an international one. The modelling work performed on the EU ozone abatement strategy and the guiding discussions in the Ad Hoc Working Group were co-ordinated with the development of the multi-pollutant protocol in the framework of the UN-ECE Convention on Long Range Transboundary Air Pollution, in which all European countries participate. Because reductions in ozone precursor emissions also significantly affect acidification, the ozone strategy had to be developed in combination with the acidification strategy.

The wide array of environmental goals and the complex way in which exceedances of environmental targets depend on emissions of SO_2, NO_x, VOCs and NH_3 made it necessary to rely on computer modelling to identify cost-effective improvements. The model system used was RAINS, an integrated assessment model which comprises databases for current and future emissions and emission control options, a module describing atmospheric transport, chemistry and deposition based on the EMEP model, and a cost-optimisation method. RAINS was used to calculate the most cost-effective combinations of emission reductions for selected combinations of environmental targets. For ozone, the environmental targets were defined as a combination of 'ceilings' (i.e. uniform EU-wide maximum ozone levels) and 'gap closures' (i.e. uniform EU-wide percentage reductions of the gap between 1990 levels and the long-term ozone objectives). After a process of trying-out, assessment and discussion, the 'central' H1 scenario was chosen, which the majority of the Steering Group viewed as a good basis for the political discussions to follow.

This scenario reduced the health-related gap by at least two-thirds and the vegetation-related gap by at least one-third everywhere. The ceilings set in the scenario formed the bases from which the target values for the ozone directive were derived. Additional calculations had to be performed to improve the correspondence of the model results with measurements from past years, though recognising that these calculations cannot be regarded as highly accurate. As a consequence the non-attainment of the target values should be assessed in a flexible and proportionate way.

The Steering Group recognised that considerable uncertainties existed in the predictions for the year 2010. In some cases they are biased or skewed: an example are the estimated costs for abatement, which are very likely on the high side because they have been based solely on "end of pipe" measures, omitting cost-saving structural changes and/or non-technical measures. The uncertainty of ozone impact on environment and health is also skewed: effect levels are known, whereas one cannot accurately estimate non-effect levels. Some members pointed to the lack of scientific confidence in determining critical levels for vegetation that are applicable to all EU countries, but at the same time most participants felt that these uncertainties essentially reflected the limitations of current scientific knowledge. Thus, one should provide for a revision, in particular, of the vegetation-related long-term objective and target value in the light of new scientific knowledge with specific account of the situation in the Mediterranean area. The following target values are proposed for ozone in 2010:

- ***Health-related target value: 20-25 days per year with exceedance of 120 µg/m³ (8h mean) as average over 3 years;***

- ***Vegetation-related target value: AOT40[3] = 8-9 ppm.hours as average over 5 years.***

In relation to existing and planned emission reductions, the new targets require further reductions of around 22% for VOCs and around 14% for NO_x. The additional cost of meeting the target values is estimated at maximum EUR 4.3 billion per year in the EU (these and the following figures refer to a scenario focusing only on achievement of the ozone targets).

A separate study was conducted to identify and estimate the benefits of these additional emission reductions. The study took detailed account of the significant uncertainties in the analysis, and tested the effects of those uncertainties on the outcome of the comparison of costs and benefits. The estimated monetary benefits of the proposed targets range from EUR 3.3 to 11 billion per year, if chronic health effects are excluded. The large range is mainly due to the uncertainty in valuation of acute mortality, where benefits derive largely from reduced damage from fine particulate matter related to ozone precursors. Other benefits are reduction of morbidity effects (EUR 1.2 billion per year), increased agricultural productivity (EUR 1.9 billion per year), materials benefits (EUR 17 million per year) and increase of production of timber and pulp (EUR 140 million per year). If chronic effects of exposure to secondary particulates on mortality are included the benefits increase by between an estimated EUR 6.1 and EUR 10 billion per year. The range again relates to variation in the method used to value mortality. Overall, the study concluded that benefits were likely to be comparable to costs or substantially higher (depending on the valuation and inclusion of mortality) though for some combinations of assumptions this would not be the case. On the other hand, for a number of effects (e.g. ecosystem damage, damage to cultural assets, direct effects of ozone precursors on heath), no reliable data exist to assess monetary benefits at all.

Strategies for the Local and Regional Scale

Ozone peaks can occur on the regional and urban scale, superimposed on the larger-scale background levels. Although EU-wide abatement will also affect local levels, it is important to address the potential for additional local measures, particularly to reduce the exposure of urban populations to ozone. It is also important to consider the potential of short-term measures, not taken into account in the EU-wide calculations.

It is very difficult to develop a generic approach, however, because local conditions differ very substantially across the EU. Ozone levels in city centres tend to be depressed compared with regional background levels due to the fast reaction of local NO emissions with ozone, though ozone plumes are known to develop downwind of cities. The effectiveness of ozone-reduction measures increases with the size of the area in which they are applied, but the scale for effective action will depend on local climatology, topography and emission characteristics, and on weather conditions during individual episodes. Consequently, there seem to be no practicable possibilities of setting a quantitative legal threshold for local actions. Since such actions are important elements in the abatement of ozone, it is proposed instead that a procedure be defined which requires Member States to explore the possibilities of local measures and, if effective local measures can be identified, to implement them.

A brief review of recent studies revealed varying scope for durable measures that could be taken on a sub-regional and urban scale in addition to large-scale measures related to the EU strategy. They can be considered part of the action programmes to be developed at Member State level to achieve compliance with target values. Such measures certainly contribute to attainment of the

[3] Calculated from 1h values from May to July, only between 8 am and 8 pm.

national emission ceilings and should be part of the requisite reporting to the Commission on abatement measures pursued by Member States.

6. REPORTING ON OZONE

It is proposed that the arrangements of the current ozone Directive 92/72/EEC for monthly and annual reporting to the Commission be maintained. It is also proposed that Member States be encouraged to provide maps of ozone over their territory, in addition to monitoring data. Exceedance of the information and general alert thresholds will trigger the release of public information on current and forecast ozone.

It is also recommended that reporting under the new ozone directive be integrated with related reports, particularly those under the National Emission Ceilings Directive. Integrated assessment of emissions and air quality will considerably improve the assessment of trends and of the effectiveness of the implemented ozone strategy.

Finally, it is proposed that a minimum set of data be defined that should be published in any other report on ozone in the EU.

1. INTRODUCTION

1.1. Background

1.1.1. Introduction

Council Directive 92/72/EEC on air pollution by ozone was adopted in 1992. It required the EU Member States to monitor ozone levels, exchange information, and inform and warn the population when high ozone levels occurred. It enabled the competent authorities of the Member States and the European Commission to obtain wider knowledge of ambient ozone, optimise the action needed to reduce ozone formation and guarantee the release of information when certain concentration thresholds were exceeded.

In 1996, Council Directive 96/92/EC on ambient air quality assessment and management (the "Framework Directive") set a general framework for EU directives on air quality. It defined the basic principles of a common strategy regarding the setting of air quality objectives, the assessment of air quality, public information and management of air quality. It listed a set of thirteen air pollutants for which the Commission should submit proposals for setting air quality objectives according to a specified time table.

In 1995 the Commission and the Member States set up the Air Quality Steering Group to help the Commission develop proposals for directives on the various air pollutants, the so-called "Daughter Directives". The Commission, the Member States and representatives of industry and non-governmental organisations participated in the Steering Group. Under its responsibility, position papers were drawn up for each pollutant, which were to serve as background documents to the Commission for the preparation of the corresponding Daughter Directive. Working Groups of the Steering Groups were set up to prepare the position papers. A proposal for the first set of pollutants (sulphur dioxide, nitrogen dioxide, particulate matter[4] and lead) was put forward by the Commission on 8 October 1997 and reached common position in the Council in September 1998. In 1996 work began on developing proposals for the second set, consisting of carbon monoxide, ozone and benzene.

A working group was also established for ozone, though concurrent developments regarding ozone meant that a more elaborate organisation structure was needed than for the other pollutants. The existing ozone Directive (92/72/EEC) required the Commission to report by no later than March 1998 on the current ozone situation and to make proposals to control ozone and to reduce precursor emissions if necessary. The negotiations on emission ceilings in the frameworks of the UN-ECE Convention on Long Range Transboundary Air Pollution and the EU acidification strategy had important links with the ozone strategy. In addition to this, the strategy development for the road transport sector in the Auto Oil I and II programmes had a strong focus on ozone. In relation to the Ozone Daughter Directive three groups were set up under the auspices of the Steering Group:

- The Ad Hoc Working group on ozone, which focused on the development of the Ozone Daughter Directive and the ozone reduction strategy,
- The Risk Assessment Subgroup, which drafted Chapter 3 on risk assessment,
- The Monitoring Subgroup, which drafted Chapter 4 on the assessment of ozone levels.

[4] The pollutants listed in the Framework Directive as Fine particulate matter such as soot and Suspended particulate matter were taken together as Particulate matter.

Annex A describes the organisational structure related to the Ozone Daughter Directive, including the participants of the various groups, in more detail.

This position paper provides a background document to support the Commission in drafting the proposal for the new Daughter Directive on ozone. Chapter 1 gives an overview of the ozone situation, taken in part from the Consolidated Report (J.P. Beck et al., 1999) produced by the European Topic Centre on Air Quality. Chapter 2 proposes how the concepts of the Framework Directive, including its special provisions for ozone, can be applied and extended in the Daughter Directive. Chapter 3 proposes objectives and threshold values for air quality, based on a review of risks and the air quality guidelines for ozone by WHO and UN-ECE. Chapter 4 makes proposals on the assessment of ozone levels in the EU by the Member States. Chapter 5 summarises the ozone abatement strategy. It is divided into a subchapter on large-scale ozone, which relates to the Commission's analysis underpinning the ozone strategy, and a subchapter dealing with the local scale, particularly urban levels. Finally, in Chapter 6, proposals are given for the reporting procedures, including specifications of the data to be reported.

1.1.2. Air pollution by ozone

In the lower layers of the atmosphere, ozone is primarily formed by a complicated series of chemical reactions, initiated by sunlight, in which nitrogen oxides (NO_x, where $NO_x = NO + NO_2$) and volatile organic compounds (VOCs) react to form ozone. These chemical reactions are not instantaneous, but take place over several hours or even days depending on the VOCs. Once ozone has been produced it may persist for several days. In consequence, ozone measured at a particular location may have arisen from VOC and NO_x emissions many hundreds or even thousands of kilometres away, and may travel further for similar distances (D. Simpson et al., 1996 and 1997, R. Guicherit and van Dop, 1977, Cox *et al.* 1975). Maximum concentrations therefore generally occur downwind of the source areas of the precursor pollutant emissions. Indeed, in urban areas, where concentrations of traffic emissions may be high, nitric oxide (NO) from exhaust emissions may react with ozone to form nitrogen dioxide (NO_2), thereby reducing ozone concentrations. However, as the air movement carries the primary pollutants away, more ozone is generated and concentrations rise in downwind areas. In some regions climatological and topographical conditions may lead to re-circulation of air masses, causing the air to reside in the region for a number of days.

VOCs are produced mainly by road traffic and the use of products containing organic solvents. NO_x is mostly emitted from transport and combustion processes. These major sources are already subject to legislative controls that will come into effect before 2010. Thus the relative importance of different emission sources will change in the future. Although NO_x and VOCs are the most important precursors of elevated levels of ozone, production of ozone can also be stimulated by carbon monoxide, methane, or other VOCs produced by plants, trees and other natural sources.

Photochemical episodes of high ozone concentrations are superimposed on a baseline which varies slightly throughout the year but annually averages around 60-80 $\mu g/m^3$ over much of Europe. This is made up partly of ozone transported from the stratosphere, and some ozone produced in the troposphere from naturally occurring and man-made precursors (in broadly equal proportions). There is evidence that the baseline has roughly doubled since the turn of the century (Volz and Kley, 1988), largely due to the increase in man-made emissions throughout the Northern Hemisphere over this period. It is unclear how baseline concentrations will develop in the future in response to global emission changes. Emissions in Asia for example are expected to increase significantly. On the other hand, and probably more importantly, emissions from Europe and North America are expected to decline.

Ozone in the troposphere is also of relevance to the climate change issue. It is currently estimated that tropospheric ozone adds 0.4 W/m^2 to the current enhanced climate forcing of 2.45 W/m^2. The latter is a result of the increase in long-lived compounds only (CO_2, CH_4, N_2O, halocarbons)

(IPCC 1995). Ozone in the stratosphere is beneficial as it acts as a screen protecting the earth from the damaging effects of incoming ultra-violet light. Ozone at lower levels, i.e. in the troposphere, will also absorb harmful UV light.

From these considerations, and bearing in mind the importance of sunlight in these reactions, elevated ozone levels occur more frequently in summer, in the southern, central and north-western parts of Europe than in the Nordic countries or eastern Europe, and in rural and suburban areas rather than in city centres. Moreover, because of the time taken for ozone to form and then be destroyed in the atmosphere, and hence the distance it can travel, the problem is an international one in many parts of Europe. Unilateral action by any one country would be of limited effectiveness in the overall reduction of ozone levels.

1.1.3. The Current Directive 92/72/EEC

Council Directive 92/72/EEC of 21 September 1992 on air pollution by ozone entered into force on 21 March 1994. In three earlier directives, air quality thresholds for the EU had been defined for sulphur dioxide, nitrogen dioxide/oxides, black smoke, suspended particulate matter and lead. In contrast to these, 92/72/EEC did not prescribe limit values for air quality, but set threshold values that triggered exchange of information between the Member States and the Commission, and the supply of information by the Member States to the public.

Purpose

The purpose of the Directive, as stated in Article 1, was to establish a harmonised procedure:

- For monitoring,
- For exchanging information,
- For information and warning the population

The aim was to enable the competent authorities of the Member States and the Commission to obtain wider knowledge of air pollution by ozone in the Community, to optimise the action needed to reduce ozone formation, and to guarantee basic public information where concentration thresholds were exceeded.

Thresholds

Table 1.1 indicates the various thresholds. These were based on assessment carried out prior to the Directive's adoption in 1992. There is now broad consensus that these values should be reconsidered in the light of more recent scientific information.

Table 1.1 Thresholds for ozone concentrations in the air, set by the current Directive 92/72/EEC

Description	***Based on***	***Value***
Population information threshold	1 hour average	180 µg/m³
Population warning threshold	1 hour average	360 µg/m³
Health protection threshold	Fixed 8 hour means (period hours 0:00-8:00, 8:00-16:00, 16:00-24:00, 12:00-20:00)	110 µg/m³
Vegetation protection threshold	1 hour average	200 µg/m³
Vegetation protection threshold	24 hour average	65 µg/m³

Monitoring

To assess ambient ozone levels and compare them with the thresholds, monitoring networks following harmonised principles had to be established. Some of the existing national networks already largely complied with the requirements of the Directive, but in some Member States major reconstruction and/or extension work had to be undertaken. Section 4.1 describes the monitoring requirements of the current Directive 92/72/EEC in more detail.

Section 1.3.3.1 gives a summary of ozone levels reported in the framework of 92/72/EEC, including an assessment of the representativeness of the current network.

Data exchange between Member States and the Commission

In the initial stage of implementation of the Directive, not all Member States were able to send the required data completely and promptly to the Commission. However, a data delivery routine has now been established in all Member States, though it has not in all cases been arranged fully according to the requirements. A routine procedure to process the data received by the Commission has also been set up. This results in a report to the Council in October, summarising the data of the preceding summer that were reported on a monthly basis to the Commission. Also in autumn a complete overview is reported of the levels of the preceding year, based on the Member States' annual reports.

Public information and warning

The Directive required Member States to issue information to the public via the media as soon as any exceedance was observed. This also stimulated the national authorities to supply the population with background information on ozone via brochures, teletext and other media, explaining the causes and the effects, and advising how to behave during episodes of high ozone. In some countries more detailed brochures were provided to doctors. Further, the Directive proved to be a stimulus to ongoing work to develop ozone forecasting systems. Some Member States were reported not to be carrying out their public information duties to the full, but in many countries the Directive has brought about a change in the public perception of the ozone problem. The population information threshold (180 µg/m³, 1-h mean) was exceeded several times per year in most Member States, while the population warning threshold (360 µg/m³, 1-h mean) was attained in a few cases. The information issued when thresholds were exceeded received considerable media attention, and increased public awareness has pushed the ozone problem higher up the political priorities list.

1.2. Emissions of precursors

1.2.1. Global emissions

In contrast to most trace gases, ozone is not emitted directly. In the troposphere, it is formed in a complex chemical mechanism from substances from anthropogenic and natural sources, in particular methane, NO_x, CO and volatile organic compounds (VOC[5]).

Table 1.2 gives an overview of anthropogenic global scale emissions (Olivier *et al.*, 1996). On a global basis it is estimated that 140 million tons/year of NO_x (expressed in NO_2-equivalents) are

[5] Because of the major role of methane in global scale ozone formation, it is customary to distinguish methane and non-methane VOC on that scale. In considerations on ozone formation on a smaller spatial scale, and also in the terminology employed in EU air quality legislation, it is more customary to use the term VOC for non-methane VOC. This convention will be applied in this position paper: VOC does not include methane.

emitted into the atmosphere (Lee *et al*., 1997), with about 60% originating from anthropogenic sources. However, over Europe anthropogenic sources are by far the dominant source of NO_x (more than 95% (Simpson *et al*., 1998)). For VOCs, total global emissions are estimated to be approximately 1 000 million tons/year (Guenther *et al*., 1995), and 90% of this amount has been attributed to natural sources while 10% comes from anthropogenic sources. Over Europe, VOC emissions are estimated at about 13 thousand tonnes from biogenic sources compared with 20 thousand tonnes from anthropogenic sources (Simpson *et al*., 1998). Uncertainties, especially regarding biogenic VOC emissions, are very large, however, with factors of 3 often quoted.

Table 1.2 Global anthropogenic emissions in 1990 (millions of tons)

	Methane	**CO**	**NO_x[1]**	**VOC[2]**
Fossil fuel (combustion)	4.8	262	72	42
Fossil fuel (production)	89			27
Biofuel	14	181	5.1	31
Industrial processes	0.8	35	4.8	34
Land use/waste treatment	211	495	20	44
Total	320	974	102	178

[1] In NO_2-equivalents

[2] In Tg C

1.2.2. EU and national emissions

1.2.2.1. Sources of ozone precursors

Figure 1 shows the trend in annual emissions of VOCs and NO_x[6] in the period from 1980 to 1995 in the EU15 Member States (Mylona 1996, Olendrzynski 1997). Emissions from biogenic sources were excluded from this inventory. The data presented are the latest officially reported emissions under the LRTAP Convention and were made available by UN-ECE. For 1990 and 1994 the data were supplemented by emission data from the CORINAIR programme (EEA-ETC/AE 1997, EEA-ET/AE,1996). This programme is run by the Member States and supported by ETC/AE (Topic Centre of Air Emissions). The CORINAIR inventory is based on data gathered by national experts in individual countries. For each pollutant it gives the contribution of individual countries to total European emissions as well as emissions per main source group, per capita and per km².
The results are estimates of actual emissions with significant uncertainties in several cases.

Figure 1.1 shows that emissions of both species increased until the late 1980s but are now decreasing. Between 1990 and 1994 VOC emissions from EU15 fell from approximately 14 000 ktonnes to 12 700 ktonnes, i.e. a reduction of 9%. In the same period, total Pan-European emissions fell by approximately 14%, demonstrating a higher decrease in central and East European countries. This may be partly due to the economic restructuring process in this part of Europe. Similar emission reductions are found for NO_x. These show a 8% decrease (from 13 500 to 12 400) between 1990 and 1994, whereas the Pan-European emissions fell by 14% over the same period. It should be noted that some of these emission trends (or lack of them) may be caused by changed methodologies being used across different years, especially for estimates prior to 1990.

[6] This section focuses on the main precursors for ozone formation on the EU scale. CO and methane, mentioned in the previous section, are more important on a global scale.

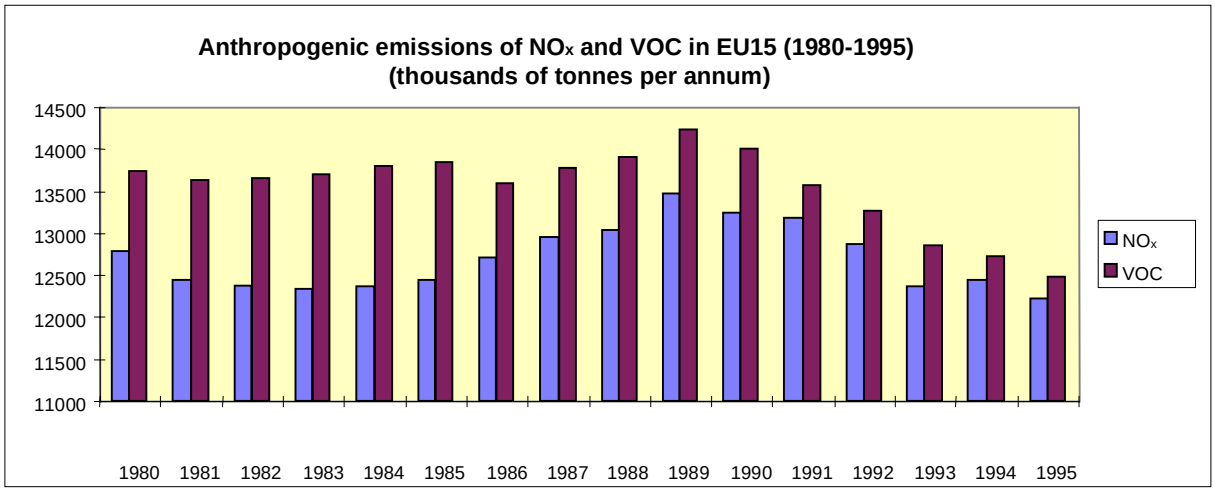

Figure 1.1 Anthropogenic emissions of NO_x (as NO_2) and VOCs (1980-1995). Source: UN-ECE / Corinair

These emission reductions can be evaluated in the framework of the 5th Environmental Action Programme (EC, 1993). The 5EAP includes emission abatement targets for the ozone source species. The VOC target is geared towards a 30% reduction by the year 2000 from the 1990 emission levels. The NO_x target also uses 1990 as its reference year, and 2000 as the year to achieve a reduction of 30%. It also aims at a stabilisation in 1994 on the 1990 levels. Figure 1.2 and 1.3 show the emission reduction achieved so far. Many countries managed to realise a significant reduction. Given the current reduction rate, it might appear unlikely that the remaining reductions of more than 20%, according to the 5EAP aim, will be met in 2000. However, the IIASA 7th Interim report, which takes into consideration legislation that is already committed to or has yet to be implemented, predicts that between 1990 and 2010 there will be an overall EU15 emission reduction of 48% for NO_x and 49% for VOCs.

Emissions from vegetation and soil contribute to hydrocarbon and NO_x concentrations in the atmosphere and therefore to ozone formation. In the EU15, averaged over the year, the contributions are in the order of 20% and 7% for VOCs and NO_x respectively. On a hot summer's day the fraction of NO_x emissions from soils may be over 25% of the emissions from combustion processes (Stohl *et al.*, 1996). As biogenic VOC emissions also increase strongly with temperature, the biogenic VOC share may, in some regions, be the major fraction of the atmospheric burden of hydrocarbons during episodes. Model calculations (Simpson, 1995) suggest that although these biogenic VOCs certainly play a role in ozone production, this is probably not sufficient to significantly affect control-decisions regarding long-term (e.g. 6-month) statistics such as mean ozone or AOT40. The main reason for this is that in many areas with major isoprene emissions the concentrations of NO_x are limiting the production of ozone. However, on shorter time scales ozone concentrations and even the results of control strategies for the peak episodes may in some cases be very sensitive to the assumed isoprene emissions. Tentative model calculations by Stohl *et al.* (1996) imply that daily ozone maxima in summer were about 8 $\mu g/m^3$ higher when soil emissions of NO_x were included. These issues are obviously important in the discussion on chemical regimes (see Section 1.3.1) and more work may be needed to determine the importance of emissions from soils and vegetation.

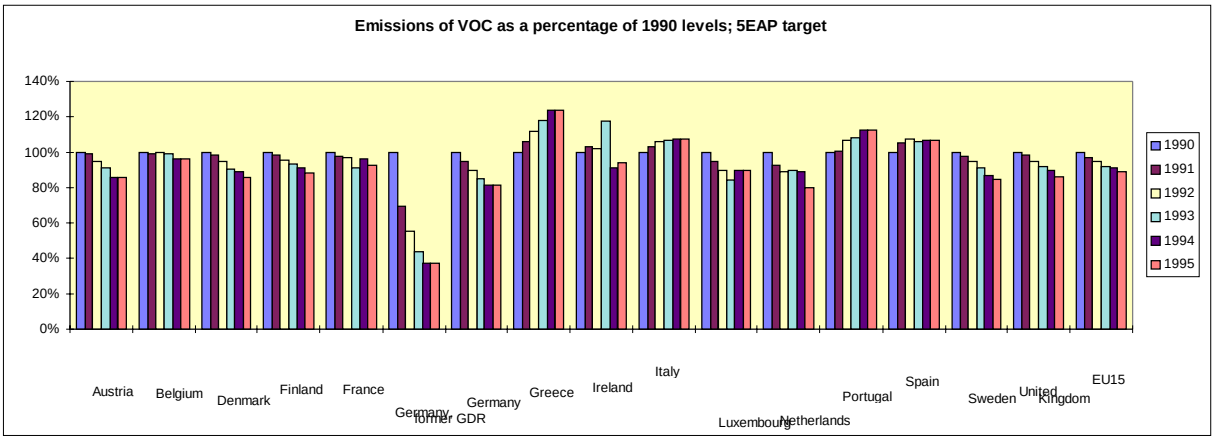

Figure 1.2 Emissions of VOCs expressed as a percentage of 1990 levels.

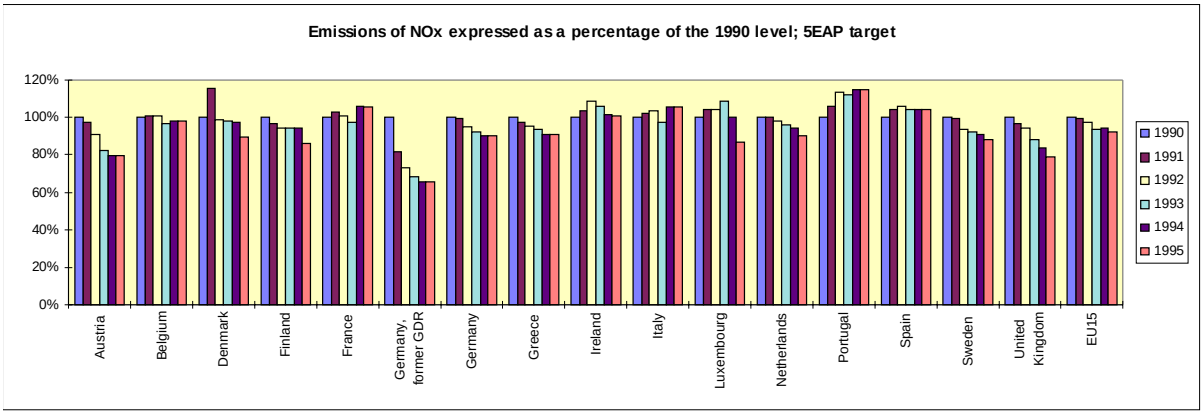

Figure 1.3 NO_x emissions expressed as a percentage of 1990 levels.

Figures 1.4 and 1.5 show 1990 emissions of VOCs and NO_x from the 5EAP source sectors. In 1990, as an average over the EU15, the transport sector accounted for 45% of total anthropogenic VOC emissions, which principally arose in urban areas. Similarly, in the case of NO_x the largest fraction originated from transport with an almost stable share of 64% between 1990 and 1994. The second largest sector is industry (approximately 35%) for VOCs, whereas for NO_x it is the energy sector, contributing about 19%. Emissions of VOCs and NO_x are declining for the transport sector overall, though increased use of motorised vehicles is partially offsetting the gains from improved car technology. It also tends to counterbalance the reductions achieved by the stationary 5EAP sectors. It seems that increased traffic intensity is currently hampering progress towards the emission reductions necessary to meet air quality objectives of O_3.

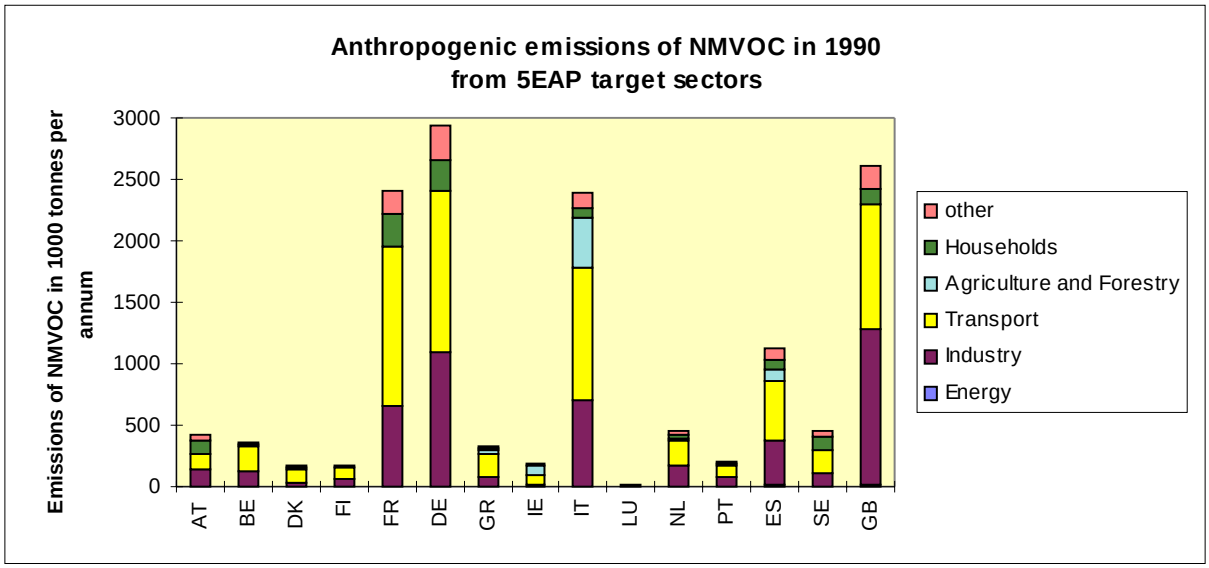

Figure 1.4 Anthropogenic emissions of VOCs per 5EAP target sector in 1990 for each Member State. Source: Corinair, EEA-ETC/AE.

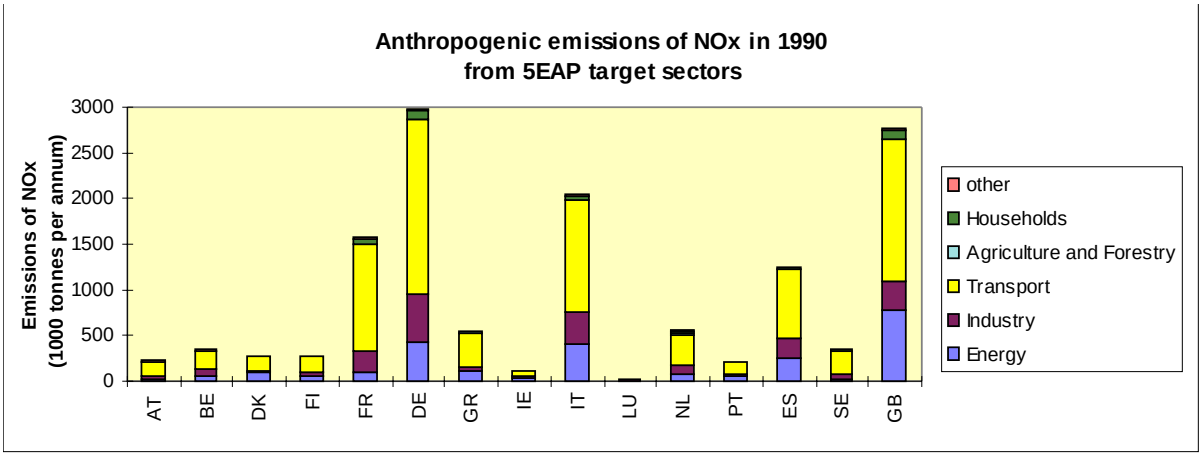

Figure 1.5 Anthropogenic emissions of NO_x per 5EAP target sector in 1990 for each Member State. Source: Corinair, EEA-ETC/AE.

Figure 1.6 shows the per capita split of the ozone source species in 1990. In this year, the consumption pattern of the average European caused the emission of 39 and 37 kg of VOCs and NO_x respectively.

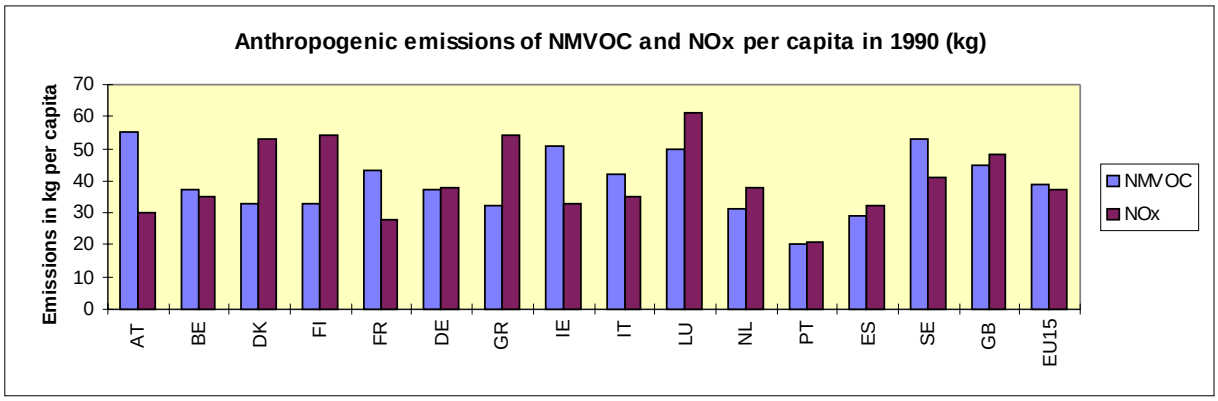

Figure 1.6 Emissions of VOCs and NO_x allotted per capita in EU15 countries. Source: Corinair, EEA-ETC/AE.

VOC and NO_x emission rates, and in particular their emission ratios, are vital parameters in determining ozone formation potential.VOCs and NO_x have different chemical atmospheric lifetimes and their concentrations change through mixing with other air masses, and deposition processes. It may be interesting to note that some Member States report a clear trend in their VOC/NO_x emission ratio between 1980 and 1995. This is probably caused by a mixture of changing volumes of activities in target sectors and varying success in VOC and NO_x emission cuts.

1.2.2.2. Emission projections

To investigate possible future developments of precursor emissions, emission scenarios for the year 2010 were developed by IIASA (Amann *et al.*, 1999). Three scenarios were constructed to reflect the likely impacts of current emission abatement policies and regulations. To allow for the 'dual track' approach adopted in Europe (regulations on emission standards for specific source categories and ceilings for national total emissions), two alternative scenarios were constructed to mimic the implications of these approaches. While the 'Current Reduction Plans' (CRP) scenario incorporated officially adopted or internationally announced ceilings on national emissions, the 'Current Legislation' (CLE) scenario relied on an inventory of (present and already accepted future) legally binding emission control legislation for the European countries. On the basis of these two scenarios a 'Reference' (REF) scenario was constructed that selected the more stringent emission ceiling for each country. In the development of the EU ozone strategy, to which the new ozone Daughter Directive is linked, the REF scenario was used as the reference line against which new emission control scenarios were compared.

Table 1.3 summarises national emissions for 1990 and those calculated for 2010 in the REF scenario. For comparison, the total emissions for European non-EU countries are given as well. For the EU-15 as a whole, the scenario calculations result in a 48% cut in NO_x and a 49% cut in VOC emissions.

Table 1.3 Emissions for NO_x and VOCs for 1990 and the Reference (REF) scenario for 2010 (Amann *et al.*, 1999)

	NO_x			VOC		
	1990	**2010 (REF)**	**Change**	**1990**	**2010 (REF)**	**Change**
Austria	192	103	-46%	352	205	-42%
Belgium	351	191	-46%	374	193	-48%
Denmark	274	128	-53%	182	85	-53%
Finland	276	152	-45%	213	110	-48%
France	1867	858	-54%	2382	1223	-49%
Germany	2662	1184	-56%	3122	1137	-64%
Greece	345	344	0%	336	267	-21%
Ireland	113	70	-38%	110	55	-50%
Italy	2037	1130	-45%	2055	1159	-44%
Luxembourg	22	10	-55%	19	7	-63%
Netherlands	542	280	-48%	490	233	-52%
Portugal	208	177	-15%	212	144	-32%
Spain	1162	847	-27%	1008	669	-34%
Sweden	338	190	-44%	511	290	-43%
UK	2839	1186	-58%	2667	1351	-49%
EU-15	**13226**	**6849**	**-48%**	**14031**	**7128**	**-49%**
Non-EU	10118	6983	-31%	7954	6635	-17%

1.3. Ozone in the troposphere

1.3.1. Atmospheric processes

1.3.1.1. Chemistry

In the polluted boundary layer, ozone is chemically formed by the oxidation process of Volatile Organic Compounds (VOCs) in the presence of NO_x and sunlight. In addition, other products are formed such as peroxy acetyl nitrate, nitric acid, aldehydes, organic acids, particulates and several short-lived radical species. VOCs act as "fuel" in the ozone formation process, whereas NO_x functions as a sort of catalyst, since it is not consumed in the formation process proper. The simplified photochemistry of ozone, which is in reality a complex and highly non-linear process, is shown in Text Box 1.

High concentrations of freshly emitted NO locally scavenge O_3, a process leading to formation of NO_2. Close to the sources this titration process can be considered an ozone sink. In addition, high NO_2 concentrations deflect the initial oxidation step of VOCs by forming other products (e.g. nitric acid), which prevents the net formation of O_3. Because of these reactions, a *decrease* in NO_x can lead to an *increase* in O_3 at low VOC/NO_x ratios, as is the case in cities. In what is often called the VOC-limited regime, therefore, emission control of organic compounds is more effective for reducing peak values of ozone pollution locally.

As an air mass moves away from an urban centre its VOC/NO_x ratio changes due to further photochemical reactions, meteorological processes and the occurrence of fresh emissions. The concentration of NO_x decreases faster than that of VOC and consequently the VOC/NO_x ratio is amplified. At high VOC/NO_x ratios occurring in background situations, the chemistry tends towards the NO_x-limited case and NO_x reductions are considered more effective for reducing ozone

in these situations. Recent work (Kramp *et al.*, 1994; Flocke *et al.*, 1994) indicates that the photochemistry in urban plumes proceeds faster than was previously assumed. The oxidation of VOCs produces more ozone over a shorter time period, and more rapid removal of NO_x. Hence the regime where ozone formation is controlled by the concentration of NO_x is reached more quickly than previously thought (Borrell *et al.*, 1995).

Text Box 1 The photochemistry of ozone formation in simplified form

	$VOC + OH + O_2$	$\rightarrow$	$RO_2 + H_2O$
	$RO_2 + NO + O_2$	$\rightarrow$	$NO_2 + HO_2 + CARB$
	$HO_2 + NO$	$\rightarrow$	$NO_2 + OH$
	$2(NO_2 + h\nu + O_2$	$\leftrightarrow$	$NO + O_3)$
net:	$(NOx+OH+) VOC + 4O_2$	$\rightarrow$	$2O_3 + CARB + H_2O$ (+NOX+OH)

VOC stands for volatile organic compound and CARB for carbonyl compounds, which play the role of hydrocarbons in further oxidation steps. OH and HO_2 are short-lived radicals which play an important role in the ozone formation process.

The complexity of the effect of NO_x emission reductions on O_3 can be illustrated by the "weekend" effect. Dumont (1996) documented that O_3 levels in Belgian conurbations were found to be significantly higher at weekends than during the week. During "smog" summers the average afternoon peak was about 20% higher on Saturdays and Sundays than on "working days". The opposite pattern occurred for NO_2; this species was lower on Saturdays and Sundays. However, the sum of O_3 and NO_2, often called Ox, was similar no matter what day was taken. The weekend effect merely exhibits a shift in the O_3/NO_2 ratio as a result of the low level of NO_x emissions during the weekend in Belgian cities (about 30% lower). Brönniman and Neu (1997) found in Swiss data two distinct patterns in the weekly cycle of ozone. When the meteorology is not favourable to ozone production, higher concentrations were observed at the weekend. However, during conditions leading to high ozone the peak on Sundays was 10-15% lower than on workdays. It is worth noting that it is *only* as a result of initial and small NO_x reductions, in the absence of concurrent reductions in VOC emissions, that the counter-acting 'weekend effect' may occur. To achieve reasonable ozone reductions, abatement of a major fraction of both NO_x and VOC emissions is necessary.

VOCs have different ozone-generating capacities. The chemical basis for these differences is now reasonably well understood. The concept of the Photochemical Ozone Creation Potential (POCP) is a widely used modelling approach to estimate the relative importance of individual VOCs for the short-term production of O_3 (Derwent and Jenkin, 1991; Simpson, 1995). POCP is defined as the change in mean O_3 when a particular species is reduced relative to the change in mean O_3 when ethene is reduced. The definition of POCP is subject to some discussion because it does not refer to: (1) the transport time scales versus the photochemical reaction time scales, (2) the levels of peroxy radicals and NO_x required during the production of O_3, and (3) the question of VOC or NO_x limitation. Where the intention is to regulate on the basis of O_3-forming potential, rather than on total mass, POCP assessments point to toluene, ethene, butane and propene as the most efficient short-term ozone producers among the most abundant VOCs. However, if one evaluates POCP values on longer time frames (e.g. 96-h), the slowly reacting alkanes become increasingly important (Andersson-Sköld *et al.*, 1992).

1.3.1.2. Transport and deposition

After emission, precursors are dispersed by wind and atmospheric turbulence. The freshly emitted pollutants mix with other pollutants, including ozone, present in background air, and a complicated

process of chemical reactions and continuous dilution takes place. During daytime, the lowest part of the atmosphere up to a few thousand metres is well mixed. At night, especially during episodes, a surface layer, typically about 100 m thick, often develops near the ground. In this shallow nocturnal layer, ozone is depleted by dry deposition on the ground and also, if near-surface emissions of NO_x are present, by titration by freshly emitted NO. In the stable layers above the surface layer, ozone and precursors remain present overnight (undergoing night-time chemical processes, and often considerable horizontal transport). The following morning, vertical atmospheric mixing is revived by the sun, causing "aged" ozone and precursors in the reservoir layers aloft to be mixed downward into the surface layer. This process of mixing down old ozone has a similar effect on the diurnal pattern of ozone concentrations as the chemical formation of new ozone.

In mountainous areas and in coastal regions in southern Europe, other important transport mechanisms add much complexity to these processes. Annex B gives a sketch of the processes at high mountain and valley sites and a description of the situation in the south of Europe, based on observations in the coastal area of Valencia, Spain. Also, ozone concentrations tend to be higher here during the day than at night.

1.3.2. Distribution of ozone and source contributions

1.3.2.1. Spatial scales

Photochemical air pollution was first thought to affect primarily urban areas. In the 1970s it was found that photochemical episodes can be associated with long-range transport of ozone and its precursors. It was also recognised that this is more often the case in northern and western Europe, where air masses are mostly advected and net transport dominates. Bouscaren (1991) concluded that in the South of Europe photochemical smog is often of a local character, but accelerated economic development appears to be changing that situation. Recent south-European studies (see also Annex B) show that ozone episodes often occur on a regional scale as a result of re-circulating air masses under conditions of weak net transport.

Ozone concentration can be enhanced in the plume of an industrial plant or in an urban plume at a typical distance of 100 km from the source. Figure 1.7 gives a schematic view of how NO_x and ozone levels change when an air mass moves over a source area of limited size such as a city. Over the source area the NO_x concentration increases and ozone is destroyed by the freshly emitted NO_x. When the air leaves the city, the NO_x concentration decreases due to dilution and loss processes, while ozone formation gradually takes over from destruction. In large source areas or when the air stagnates or re-circulates in an area, formation may already outweigh destruction in the source area.

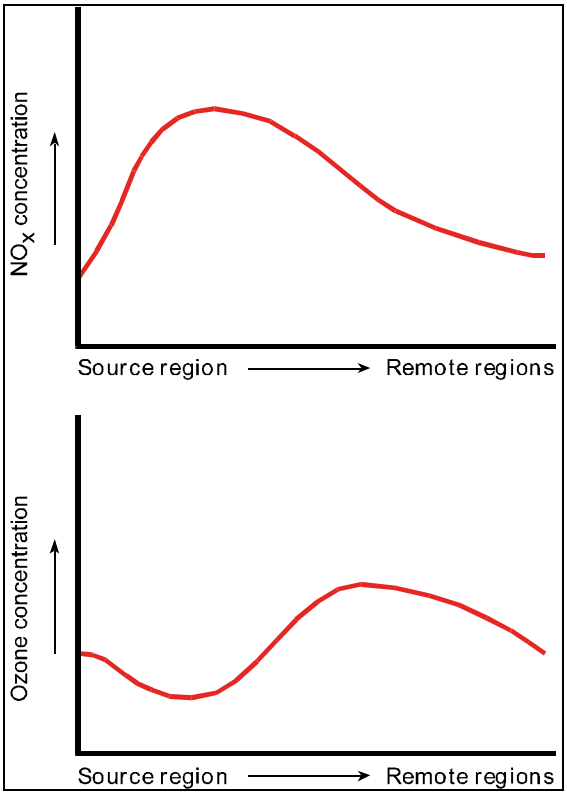

Figure 1.7 Schematic view of the development of NO_x and ozone levels in air passing over a source region

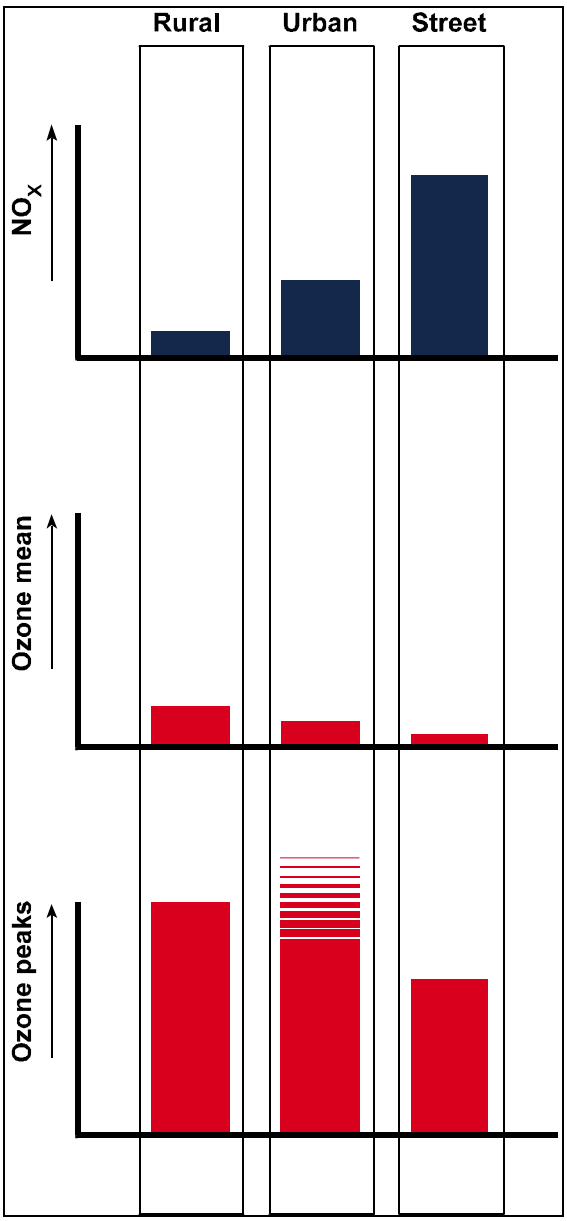

Figure 1.8 Qualitative comparison of NO_x and ozone levels in rural, urban and street environments

For an overview of ozone phenomenology, it is instructive to distinguish between spatial scales. Such schematisation also relates to the EU reduction strategy. Figure 1.8 gives a comparison of typical levels for ozone and NO_x on the rural, urban and street scale. NO_x behaviour is easy to understand: close to sources the concentrations are high, while low NO_x concentrations prevail in rural areas. The annual averaged ozone levels in the atmosphere exhibit a reversed pattern: ozone concentrations are typically higher in remote areas than close to sources. This behaviour can be understood from the discussion in Section 1.3.1.1: under "average" conditions, NO_x levels are often high enough for ozone destruction to be the dominating process. During photochemical episodes, however, time scales for atmospheric chemistry are quite different and the concentration pattern changes accordingly. Ozone concentrations in a city can then be higher or lower than the rural levels around it, depending on conditions like weather, topography, city size, etc. In the south of Europe the conditions seem to be more favourable for net ozone formation to occur within the fringes of large, but confined airsheds (Milan, Athens, Madrid) than elsewhere in Europe. In streets, the NO_x levels are high enough to depress ozone concentrations even during episodes.

Figure 1.8 does not illustrate the essential role of VOCs. Although at very high concentrations of highly reactive VOCs, ozone destruction may locally be of importance, VOCs are in practice not associated with ozone destruction. VOC species differ greatly in reactivity (see also Section 1.3.1.1), and the scale (the typical distance from the source) on which they give rise to ozone formation varies accordingly. The least reactive volatile organic compound is methane; due to the low time scale of its reactivity, methane is mainly relevant for global-scale ozone formation. Moderately reactive VOCs have their main impact on the regional (and continental) scale. The more reactive ones can cause ozone formation on the urban scale, including industrial areas and surroundings. In streets, the residence time of pollutants is too short for VOCs to affect ozone levels.

Large-scale ozone in Europe

Grennfelt *et al*. (1987; 1988) and Feister and Pedersen (1989) were the first to report summer ozone levels in Europe as showing an increasing gradient from the Northwest to the Southeast part of the OXIDATE network. Unfortunately, their analysis did not extend much south of the Alps due to the limited availability of data from the south and east of Europe. Later reports (NILU/CCC 1990 - 96) confirmed the gradient pattern in ozone.

A quantitative estimate of ozone in summer and winter was provided by Beck and Grennfelt (1994). Based on measurements from 68 rural and background stations they found that the average diurnal maximum in summer ranged from 60-80 $\mu g/m^3$ in the north-western part to 120-140 $\mu g/m^3$ in the central part of Europe. Figure 1.9 illustrates a modelled version of the gradient in the averaged diurnal maximum ozone concentration during summer over Europe (Simpson *et al.*, 1997). The European marine boundary layer background concentration, i.e. the concentration in air advected from the Atlantic, was established at 60-65 $\mu g/m^3$ (Borrell *et al.*, 1995). It may be useful to note that not enough information has been reported in the framework of the Directive for this general picture on rural ozone to be compiled. Measured data from the Eurotrac-TOR and EMEP networks and EMEP model activities were used.

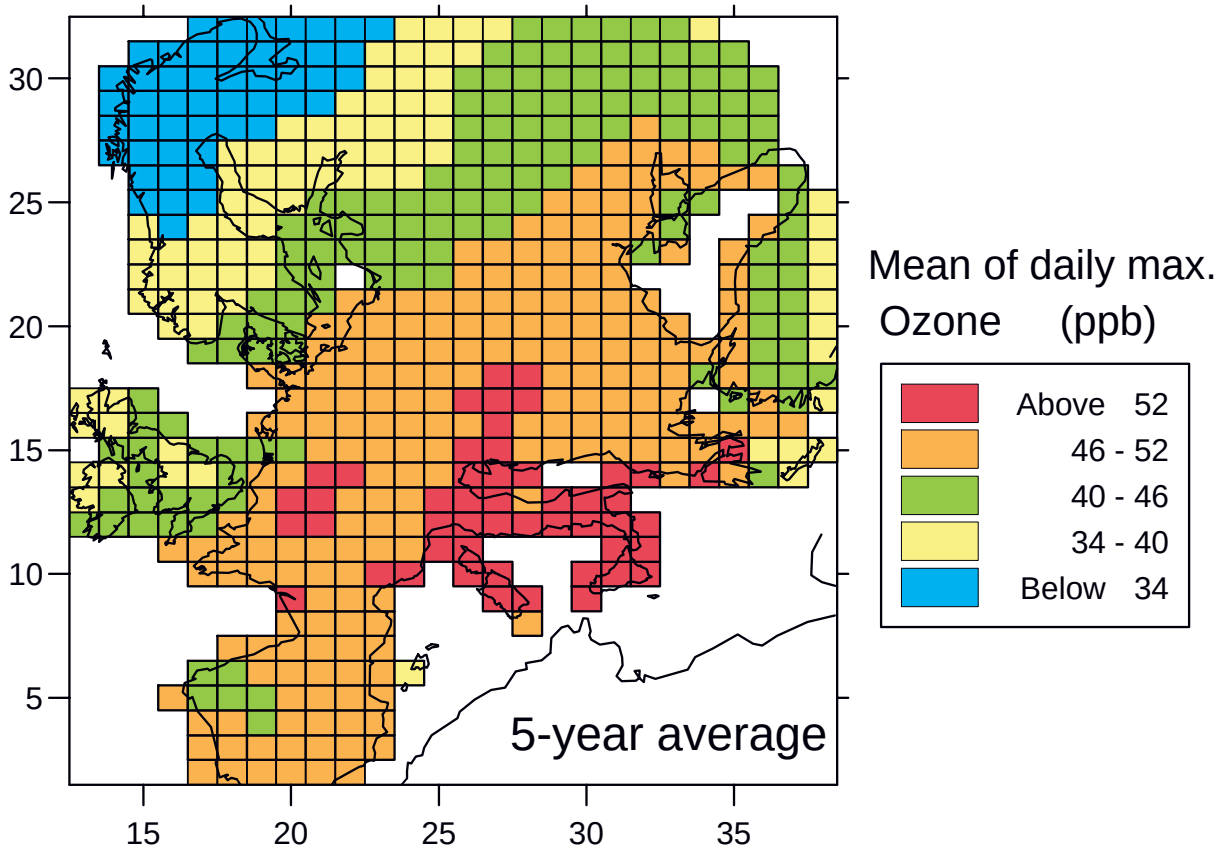

Figure 1.9 Modelled 5-year mean of daily summer maximum concentration of ozone. The calculation was performed using constant emissions at the 1990 level and meteorology from 5 summers (1989, '90, '92, '93 and '94). 1 ppb $O_3 \approx 2$ µg/m³. Source: Simpson *et al.*, 1997.

The seasonal variation in ozone, with a broad summer maximum and a winter minimum, is observed at many individual sites on the continent. On a seasonal basis, atmospheric processes in the polluted European boundary layer superimpose 30-40% on the boundary layer background concentration in summer. Most rural inland stations show a typical diurnal pattern as a mean for the summer months, with a minimum in the morning and a maximum in the afternoon. The decrease during the night and early morning is caused by dry deposition. After sunrise, photochemical formation from precursors begins and gradually grows as the amount of sunlight increases. The mixing of air from layers aloft and from the free troposphere also plays a role. Unpolluted elevated stations often show a less pronounced diurnal variation due to the small influence of dry deposition, and advection of homogeneous and relatively undisturbed air.

Regional and urban scale ozone

It is very difficult to give a comprehensive picture of regional and urban scale ozone. Under most meteorological conditions ozone formation from local emissions is more than compensated by depletion of ozone by local NO_x emissions, but under episodic conditions the ratio between formation and depletion changes markedly. Since this ratio is highly dependent on precursor concentrations, which often vary greatly across a city or region, it is difficult to predict whether the net result will be formation or depletion. In city centres depletion almost always prevails, but when an air mass remains over a city for some time, ozone accumulated during several days of formation may tip the balance. In suburbs and further downwind of large cities, where local NO_x emission rates are lower, ozone levels can be considerably elevated compared with rural background levels. Typically the build-up of ozone in urban 'plumes' takes several hours; as a result of the interplay between formation and dispersion, the highest levels in such plumes occur at distances of up to about 100 km downwind. Peak concentrations reported were between 10% and more than 50% higher than levels outside the plume.

A significant difference is believed to exist between cities in northern/central and southern Europe, although data are scarce, especially on southern cities (see also Annex B). During episodes, the levels in cities in northern and central Europe can be considered to be part of a large area of high ozone, and topographic conditions do not usually cause air to remain very long over a city. In the south, human activities tend to be concentrated in more or less isolated urbanised areas. These areas are often also enclosed by mountains. During stagnant conditions, the air can be trapped by surrounding mountains or re-circulated in a local land-sea breeze system. Under those conditions, background levels tend to be less important and high local ozone production is more likely.

1.3.2.2. Temporal scales

To understand ozone phenomenology, it is instructive to distinguish the (annual, seasonal) average levels from the peak levels that occur during photochemical episodes. Much research on large scale ozone, e.g. trend analyses, has focused on long-term averages. From the viewpoint of the development of the ozone Directive, however, the peak values are more important, since air quality objectives will be expressed in terms of exceedances of thresholds (see Chapter 3).

Long-term average ozone

During most of the year, photochemical formation of ozone proceeds much more slowly than during episodes. In regions where substantial human activity takes place, ozone depletion by NO_x outweighs formation, and ozone tends to be lower than in remote areas. Compared with episodes, ground level concentrations are determined to a higher degree by ozone of free-tropospheric or stratospheric origin.

Episodes

Episodes of increased ozone occur over most parts of Europe every summer. During these episodes ozone concentrations rise to several times the boundary layer background over large areas of Europe. Meteorology plays a very important role in the formation of episodes, which vary across Europe. In very general terms summer episodes in northern and central Europe occur under anticyclonic stagnation, which produces short-lived but intense episodes, lasting around one week. Southern Europe falls within the sub-tropical latitudes and is under conditions of weak anticyclonic subsidence in summer. In these conditions, air masses can re-circulate without moving away from the region, and episodes can last for prolonged periods.

1.3.3. Observed concentrations

1.3.3.1. Exceedance of the Directive 92/72/EEC thresholds

The Consolidated Report gives an overview of measured concentrations reported to the Commission in the framework of the current Directive 92/72/EEC on air pollution by ozone (EC1992a). This overview is based on more detailed annual reports issued by the European Commission (e.g. de Leeuw et al, 1997 and references to section 4) and EEA (see http://www.eea.eu.int/frdocu.htm). The following overview of exceedances of the thresholds set in the current Directive (see Table 1.1) was taken from the summary of the Consolidated Report. N.B. The thresholds for the protection of human health and vegetation differ from the revised WHO guidelines on which the air quality objectives proposed in this position paper are based.

- The threshold for the protection of human health is exceeded substantially and in all Member States. More than 90% of the 41 million citizens living within a 10 km radius of an urban/street monitoring station were exposed to a threshold exceedance at least once in 1995, and more than 80% of them were exposed to >25 days of exceedance. If we assume that the ozone climatology observed by the current network can be extrapolated to the full EU15 population, then approximately 330 million people (94%) may be exposed to at least one exceedance per year.

- In 1994 and '95 both the 24-h and 1-h averaged thresholds for the protection of vegetation were exceeded substantially, in all Member States and frequently. In 1995, the full EU15 area of coniferous forest and arable land experienced exceedances of the 24-h mean threshold. In less than 1% of the area of broad-leaved forest were exceedances not observed. The guideline for crops and semi-natural vegetation of 3 ppm.h is exceeded in all Member States except Finland. As an average over all EU15 countries only 6% of arable land is not exposed to exceedances of the guideline. In the case of forests, Scandinavia, Ireland and the United Kingdom are almost fully free from exceedances. Forests elsewhere in Europe experience exceedances of a factor 2~3 of the guideline.

- The threshold value for providing information to the public is exceeded in almost all Member States every year. An estimate from the 1996 data revealed that this concerned about 31 million Europeans, which approximates to 45% of the urban population living in cities with operational monitoring in that year. The warning threshold is reached occasionally, in particular in the Southern countries.

Tentative estimates of the spatial coverage of the 1995 ozone monitoring network indicate that it is insufficient to appraise the ozone situation over all Member States. The current subset of rural/background stations in the EU countries is estimated to cover a maximum of 20-40% of forests and 30-50% of crops, both depending on the chosen radius of representativeness of the observatories. Even if a radius of 100 km is used, the coverage of arable land is below 25% for 4 Member States. This number increases to 8 countries if a 50 km radius is used. The subset of urban/street sites covers no more than 12% of all EU15 residents and approximately 25% of EU city dwellers (city defined as > 50 000 inhabitants).

1.3.3.2. Exceedances of the WHO guidelines

Section 3.3 in Chapter 3 gives an overview of exceedances of the WHO guidelines for ozone in the EU.

1.3.3.3. Trend, inter-annual variability

This section summarises the ozone trends observed in the past. Future developments, which are dependent on economic growth and emission reduction measures, will be addressed in Chapter 5 and references given there.

The first quantitative measurements of the O_3 mixing ratio in Europe were made at the Observatoire de Montsouris near Paris, between 1876 and 1886. The 24-h averaged concentration was then about 20 µg/m³ (Volz and Kley, 1988). It may be interesting to note that these data (1000 samples) show exceedances of the current EU threshold value for the protection of vegetation (65 µg/m³ 24-h average) during somewhat less than 1% of all observations. (Volz-Thomas, pers. comm.). Most of these occasions arose in February and one occurred in May and they reflect the influence of air with a free tropospheric origin reaching ground level.

In the 1950s the 24-h mean rural ozone levels had increased to 30-40 µg/m³ and rose further to reach 60 µg/m³ by the 1980s (Feister and Warmbt, 1987). Today, daily mean concentrations are at least a factor of 2 higher than in the pre-industrial era (Borrell *et al.*, 1995; Staehelin *et al.*, 1994). Over the last decade, little or no ozone increase has occurred as a general pattern over rural Europe. Concentrations have decreased somewhat in the Netherlands (Roemer, 1996), while in the south of Germany several sites report a 2% annual upward trend (Scheel *et al.*, 1997). At the Irish coastal fringe of the Atlantic an upward trend of about 1% per year during summer in polluted air from Europe is observed (Simmonds, 1993).

Little work has been done on deriving trends in the occurrence of *ozone episodes* in Europe. Since extreme values are more subject to fluctuations than annual averages, trends in ozone peak values tend to be masked by the inter-annual variation caused by meteorological fluctuations over the

years. However, some countries reported data on the 98th percentile covering the period 1989-1996 in the framework of the ozone Directive. This database indicates a significant upward trend in the 98th percentile of a few µg/m³ at 2 sites in Belgium and Luxembourg, whereas a significant downward trend over 1989-1996 is observed at 18 stations in the United Kingdom and the Netherlands. There was no evidence for any trend at 35 stations in these four countries (de Leeuw and van Zantvoort, 1997). No clear picture for a country or region emerged. A word of caution is indispensable here. Collocated data on NO_x were not available, which prevents us checking whether the observed trends are caused or masked by changes in NO_x concentrations. We recommend using the sum of NO_2 and O_3, often called Ox, as a parameter to overcome the interfering influence of the titration effect, since Ox is insensitive to titration (Guicherit, 1988). Furthermore, changes in measurement techniques or operation procedures may bias the detection of trends (Roemer, 1997).

The odd historical record, derived from semi-quantitative measurements only, is available for ozone in the *urban* environment. Annual ozone averages of 40-60 µg/m³ were reported for Athens in the first two decades of this century (Cartalis and Varotsos, 1994). Similar levels in the 1890s were documented for Zagreb (Lisac and Grubisic, 1991).

The annual 98th percentile of ozone in Central London varied between 60 and 140 µg/m³ and showed a significant trend of -2.8 µg/m³ per year between 1973 and 1992 (PORG, 1987; Bower *et al.*, 1991). Basic ozone statistics from several other north-west European urban stations show values in comparable ranges over the last 5 to 10 years. A record from a suburban station of Athens (Liosia) shows that the monthly mean concentration exhibited an average increase of about 15% per annum in the period 1984-89. In 1987 the *monthly mean* values started to exceed 110 µg/m³ (Moussiopoulos, 1994). Note that this value represents the current EU *8-h averaged* threshold value for the protection of human health. In 1988 this threshold was exceeded on 140 days at this monitoring station.

The identification of a trend in ozone *episodes* in the urban environment may be more important. Table 1.4 presents the number of exceedances of the 110 µg/m³ 8-h averaged concentration (12-20 h) at the urban sites represented in the AIRBASE database. Since data from only six countries are included, the representativeness of the table for the EU is not clear. The table shows exceedances to occur at all urban sites. Over the years available, no significant trend can be detected. The year-by-year meteorological variation is likely to be the major cause of the large inter-annual variation.

Inter-annual variation is also very important for assessing the future development of threshold exceedances. Due to this variability, the effect of emission reductions on ozone peaks cannot be observed in a period of just a few years. Chapter 4 analyses inter-annual variability in relation to monitoring requirements. These fluctuations should also be taken into consideration in the setting of ozone air quality objectives.

Table 1.4 The number of exceedances of the 110 µg/m^3, 8-h average threshold value at several (sub)urban sites in the period 1982 - 1995

Country	Station	City	1982	1983	1984	1985	1986	1987	1988	1989	1990	1991	1992	1993	1994	1995
Belgium	I.R.M. Av. Circulaire	Brussels	*	*	*	*	10	10	4	37	23	11	18	21	34	42
Belgium	St-Kruiswinkel	Ghent	*	*	*	10	8	14	7	16	33	7	21	9	20	21
Belgium	Namur Ville en Waret-Vezin	Background stations	*	*	*	*	6	10	12	39	41	0	11	9	31	32
Greece	Patission 147	Athens	*	0	0	*	*	10	17	17	14	1	*	*	*	*
Greece	Smyrni Cementery of N Smyrni	Athens	*	*	*	*	*	20	48	60	34	71	*	*	*	*
Greece	Aspropyrgos	Athens	*	23	9	*	*	*	*	*	*	*	*	*	*	*
Greece	Pireas Platia Dimotikou Theatrou	Athens	*	*	*	*	*	*	59	83	30	8	*	*	*	*
Netherlands	Florapark	Amsterdam	*	*	*	*	7	9	8	27	30	6	19	9	14	20
Netherlands	Const.Rebecqueplein	The Hague	*	*	*	*	0	11	8	28	37	17	23	9	20	19
Netherlands	Schiedamsevest	Rotterdam	*	*	*	*	8	9	7	32	20	11	20	*	*	*
Netherlands	Kard. De Jongweg	Utrecht	*	*	*	*	*	0	10	24	17	1	10	8	17	20
Netherlands	Witte Vrouwenstraat	Utrecht	*	*	*	*	*	0	0	5	4	*	4	*	*	*
Netherlands	Tuin Utrechtse Bibliotheek	Utrecht	*	*	*	*	*	0	4	12	24	9	24	4	17	32
Netherlands	Amsterdamse poort	Haarlem	*	*	*	*	18	4	7	3	*	*	*	*	*	*
Netherlands	Keizer Karelplein	Nijmegen	*	*	*	*	1	6	2	*	*	*	*	*	*	*
Netherlands	Arnhemseweg	Apeldoorn	*	*	*	*	*	*	*	*	33	9	20	8	22	17
Netherlands	Floreslaan	Vlaardingen	*	*	*	*	3	5	6	25	13	*	9	4	15	19
Portugal	Lisboa Bairro Alto - R. do Seculo 51	Lisbon	*	*	*	*	*	*	4	6	1	1	2	*	*	*
Portugal	Montes Chaos	Sines	*	*	*	*	2	2	13	88	4	0	10	*	*	*
Spain	Plaza Castilla - Avenida Castellana	Madrid	*	*	*	*	*	*	*	*	*	*	*	1	*	*
Spain	Poble Nov - Pl.Doctor Trueta	Barcelona	*	*	*	*	*	30	39	8	1	13	16	*	*	*
Spain	Molina Pl.	Barcelona	*	*	*	*	*	11	20	16	0	5	9	1	*	*
Spain	Montcada I Reixach	Barcelona	*	*	*	*	*	0	0	*	*	0	2	*	*	*
Great Britain	Central London Lab-Minster House	Greater London	0	0	0	15	8	1	0	13	2	*	*	*	*	*
Great Britain	Bridge Place	Greater London	*	*	*	*	*	*	*	*	4	0	0	*	*	0
Great Britain	Stevenage - WSL	Stevenage	0	23	31	10	9	4	2	*	*	*	*	*	*	*

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Volz, A. and D. Kley (1988) “Evaluation of the Montsouris Series of Ozone Measurements made in the Nineteenth Century” Nature 332, 240-2. Basic Legislative Concept For An Ozone Daughter Directive

2. BASIC LEGISLATIVE CONCEPT FOR AN OZONE DAUGHTER DIRECTIVE

2.1. Introduction

This chapter outlines a number of basic considerations relevant for structuring the Daughter Directive for ozone, taking into account the constraints set by existing Air Quality legislation (Council Directive 96/62/EC & Council Directive 92/72/EEC), environment policy programmes and abatement proposals, as well as the specific nature of ground level ozone as a secondary pollutant.

The considerations are based on the following general principles:

(1) **Consistency** between the abatement strategy for ozone and the strategy approach already adopted for acidification (COM(97)88 final) and the ongoing and future activities under the UN-ECE CLRTAP[7].

(2) **Coherence** between the abatement strategy for ozone and the Ozone Daughter Directive, which will accompany the strategy and contain matching air quality standards.

(3) **Appropriate embedding** of the Daughter Directive in the Framework given by the Air Quality Framework Directive.

(4) **Transparency** of new ozone legislation, which means embodying the objective set by the Fifth Action Programme (OJ C 138/1) of achieving the purely effect-based WHO levels in the long term. Given the difficulty of complying with this objective throughout the Community, transparency requires that an interim objective be set which takes account of feasibility and cost-effectiveness.

2.2. Potentially relevant definitions in the Framework Directive

The following section describes briefly some definitions in the FWD which might be relevant when producing daughter legislation for ozone.

2.2.1. The Limit Value

The Framework Directive defines a limit value as:

> *"a level fixed on the basis of scientific knowledge, with the aim of avoiding, preventing or reducing harmful effects on human health and/or the environment as a whole, to be attained within a given period and not to be exceeded once attained."*

When a limit value is exceeded, Member States are obliged to assess the pollution in the zone concerned, including the sources causing the exceedance, to plan measures to prevent further exceedance, and to report this to the Commission. Air Quality assessment is still required where there are no exceedances, though the requirement will be less stringent depending on the ratio between the limit value and the existing pollution load.

For the sake of coordination, an attainment period has to be defined within which the implemented abatement measures must bring about compliance with the Limit Value in the Member States.

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[7] Convention on Long Range Transboundary Air Pollution.

From a juridical point of view the limit value imposes a legal commitment on Member States to ensure attainment without derogation.

2.2.2. The Margin of Tolerance

The Framework Directive defines a margin of tolerance (MOT) as:

> *"the percentage of the limit value by which this value may be exceeded subject to the conditions laid down in this Directive."*

- Where the limit value plus the MOT is exceeded, the action required is the same as for exceedance of a limit value that has no MOT.
- Where the limit value is not exceeded, the MOT has no relevance.
- Where the limit value is exceeded, but the MOT is not exceeded, Member States must take the requisite measures to ensure compliance with the limit value (Article 7(1)). Thus the MOT does not affect the obligation to take measures. However, Member States are in this case not obliged to inform the Commission of the measures. They will only report on exceedances in a certain zone.

2.2.3. The Target Value

The Framework Directive defines a target value (TV) as:

> *"a level fixed with the aim of avoiding more long-term harmful[8] effects on human health and/or the environment as a whole, to be attained where possible over a given period."*

As with the limit value, an attainment period must be set during which the target value should be reached. The insertion "*as far as possible"* indicates the difference between the target value and the limit value. The limit value definition does not allow any leeway concerning the compliance obligation, whereas adding "*as far as possible"* and omitting the phrase '*..not to be exceeded once attained"* in the definition of the target value provides for such circumstances as

- transboundary ozone transport
- strongly varying meteorological conditions

which might lead to unavoidable or unforeseen exceedances of the target value, even though the Member States concerned may have implemented adequate abatement measures.

The Framework Directive explicitly mentions the possibility of setting target values for ozone in addition to or instead of limit values.

Article 4(1) describes the consequences of target value exceedances as follows:

> "*If a target value […] is exceeded, Member States shall inform the Commission of the measures taken in order to attain that value. On the basis of this information the Commission shall evaluate whether additional measures are necessary at Community level and, should the need arise, shall submit proposals to the Council.*"

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[8] The French text of the Directive, which served as the source for the other language versions, has a somewhat different meaning. The following is a more appropriate English translation:

"The target value shall mean a level fixed with the aim in the long term of avoiding harmful effects on human health , to be attained as far as possible in a given period."

The connection between assessment requirements and approximation of limit values is not made explicitly for target values, because the FWD makes no further reference to the target value.

2.2.4. Upper and Lower Assessment Level

Article 6 of the Framework Directive prescribes what assessment methods to apply. It stipulates that in "agglomerations" (zones which have a special status in the Framework Directive) measurements are always mandatory if an alert threshold has been set. It also links three assessment regimes to two threshold levels (Upper and Lower Assessment Levels; UAL/LAL) below the limit value, which serve as criteria to distinguish between these regimes. Exceedance of the UAL or LAL determines which assessment regime is prescribed in the zone, though it has no implications for air quality management.

The Framework Directive makes several prescriptions regarding these three types of zone. Table 2.1 indicates the assessment regimes associated with these types.

Table 2.1 Summary of Assessment Regimes

Zone	Assessment regime, from the strictest (top) to the most relaxed (bottom) requirements
Where highest levels > UAL	Based on continuous measurements (at least one site per zone), may be supplemented by modelling
Where highest levels > LAL	Combination of continuous measurement (at least one site per zone) and modelling allowed
Where highest levels < LAL	Modelling, objective estimation, indicative measurements allowed

The UAL and LAL will be chosen on the basis of the interannual variation in the concentrations derived from empirical data. The approach used in position papers for other pollutants suggested setting the UAL or LAL below the limit value at two or three times the standard deviation, respectively. The accuracy of the least accurate assessment methodologies in regimes 2 and 3 should be sufficient to conclude, when concentrations are found to be below the UAL/LAL, that the limit value is not exceeded in reality.

In chapter 4.2 it is proposed not to utilise this concept, but instead to relate the assessment requirements to the long-term objective.

2.3. Proposal of a two-level objective for ozone

In the following approach a two-level objective is proposed which recognises two environmental goals pertaining to ozone,

- A **long-term objective** and
- The **target value** serving as an interim objective.

The aim is not only to be coherent with the existing frame of the Air Quality Framework Directive, but also to provide the ozone Daughter Directive with a reasonable structure that takes account of the special nature of ozone pollution.

The **long-term objective** reflects the clearly stated commitment in the 5th Environmental Action Programme that future EU air quality legislation would be based on WHO guidelines. This objective is based exclusively on consideration of the effect and impact of ozone on human health and the environment.

The interim objective expressed through the **target value** takes into account additional considerations of practicability, feasibility and costs. According to the Framework Directive this ozone level should be attained as far as possible *within a given period*. The reason for proposing an interim level lies in the large difference between the level of the long-term objective and the current ozone load, reflected in the frequent exceedances of the current protection thresholds in almost all EU regions. Scenario calculations in the analysis performed to devise the ozone strategy showed that even with a "Maximum Technical Feasibility Reduction" scenario (assuming technical measures only), the long-term objective is not achievable within a foreseeable time horizon. It therefore seems difficult at this stage to associate the target value with the WHO levels while defining an attainment period for it based on realistic assumptions. Furthermore, to ensure coherence with the emission reduction objective which will be established when setting national emission ceilings for relevant precursors, the target value will be a reflection of this objective in terms of the corresponding pollution level.

In the scientific analysis for the strategy development, the interim objective for ozone was defined as an equal relative improvement everywhere ("gap-closure"), while the additional criteria to meet a uniform absolute level ("ceiling") must also be fulfilled, which emphasises reductions where the highest regional ozone pollution occurs. Given the legal difficulty of defining the target value as a relative standard, it is proposed that a uniform, absolute value be used for the Target Value, derived from the "ceiling"-type objective chosen in the ozone strategy.

In developing the ozone strategy, a variety of absolute levels as well as gap-closures have been investigated with regard to their feasibility, costs and environmental benefits, providing an objective basis for deriving a target value expressed as an absolute level.

To be compatible with the strategy, and for reasons of transparency, the type (e.g. AOT40) of threshold used for the long-term objective should also be applied for the Target Value, then allowing a certain number of exceedances or additional accumulations of the LTO, respectively.

In principle the interim objective could also be defined as a Limit Value within the terms of the Air Quality Framework Directive. However, due particularly to the transboundary nature of the ozone problem and the marked meteorologically induced variability, the strict compliance obligation for single Member States imposed by the Limit Value is hardly reasonable. It is therefore proposed that the **Target Value** definition, which offers some leeway in this respect, **be used as the interim objective**.

Finally, the Daughter Directive should contain a provision for review and revision of the Target Value, the goal always remaining achievement of the long-term objective. This would provide a reference point for all future work, following the intention of the 5th Environment Action Programme.

The explicit introduction of the **long-term objective** into the Daughter Directive would ensure that

- The undertaking of the 5th Environmental Action Programme to *guarantee levels of Air Quality which are not detrimental to health and environment* will be maintained. These critical levels for air pollution were defined by the guidelines of the WHO.

- A rational basis is given for the definition of assessment requirements laid down in the Air Quality Framework Directive.

- The principle adopted for the existing ozone Directive, to set thresholds corresponding to the WHO guidelines, would be maintained in the Daughter Directive.

Consequently, the long-term objective is also being used to define the environmental objectives for ozone in the ozone strategy, which is being developed simultaneously. It marks the "gap" relative to the situation in 1990, which the interim objective is designed to "close" as far as possible.

Because of the above, the long-term objectives should be directly based on the WHO air quality guidelines (see Chapter 3.2). Preferably, they should be equal to the WHO guidelines, but for operational reasons it will be necessary to slightly refine or redefine the guideline levels in terms of equivalent thresholds that can be readily assessed in a measuring network.

Scientific knowledge of the risks ozone poses to health and vegetation has expanded and improved considerably in the last few years, and this process is expected to continue in the years ahead. Under Article 4(2) of the Framework Directive the Commission is responsible for re-examining the scientific basis for the air quality limit values and alert thresholds. It is proposed to require that this review be undertaken and followed by any proposal for revision of any of the ozone air quality thresholds that the Commission deems appropriate within five years. Reviews should also consider the feasibility of setting a target year for attainment of the long-term objective.

Given the strong link between revising the target value and further emission reductions, a review of control strategies including emission reduction targets is also deemed necessary. In updating the ozone strategy one should also take into account future requirements of abating acidification and improving air quality, especially particulate concentrations.

Alternative Viewpoints

Two opposite viewpoints were raised during the discussion in the working group, each proposing an approach based only on one objective.

The role of the long-term objective was queried because to set the long-term objective at the WHO guideline level would make it apparently unachievable in a foreseeable time frame. It would be more reasonable to base legally binding measures on an attainable target which brought tangible health and environmental benefits without imposing disproportionately high abatement costs. While a target value should be established which allowed for feasibility and costs, the long-term objective should be abandoned.

There was a general feeling, however, that the spirit of the 5th Environmental Action Programme required embodiment of the WHO levels in air quality legislation, and that current knowledge did not justify abandoning the link between the WHO levels and the respective standards already established in the current Ozone Directive.

A different approach was favoured of setting only one objective at WHO level, termed target value according to the Framework Directive. The role of this target value would be basically identical to the long-term objective proposed above. It did not seem necessary to establish an interim objective called "target value" as a sort of trigger for abatement measures in the ozone Directive, for the following reasons: (1) an interim objective for ozone was set anyway in the analysis which was used as a basis for deriving concrete emission reduction targets within the ozone strategy; (2) the uncertainty of the model predictions made it unrealistic to expect individual Member States to take further measures before 2010, the deadline for attaining the target value; (3) the transboundary nature of the ozone problem meant that target value exceedances could not be avoided by measures taken by individual Member States, and that co-ordinated European action would be needed instead; (4) emission reduction measures needed for compliance with the emission ceilings are based on a cost-effective strategy, so that further measures taken by individual Member States would not be cost-effective. Setting a target value at WHO level at a later date than 2010 would stress the need for co-ordinated European action because this level is exceeded in the whole Community.

However, given the abovementioned definition of the target value in the Framework Directive, which required an attainment period to be set, and in view of the difficulties of ensuring compliance within a foreseeable period everywhere in Europe, it was generally felt most reasonable to use the target value as an interim objective. Allocating a realistic time period within which attainment is aimed at (i.e. identical to the attainment of the emission ceilings) and deriving the target value from the integrated assessment analysis would make the target value a benchmark for the success of the abatement strategy. The main aim is to identify a potential need to review the abatement strategy at EU-level. For those regions where transboundary effects seem the prevailing factor, the target value will probably not trigger further measures, since large-scale transboundary features should be fairly well covered by the integrated assessment modelling. Mainly in southern European states, where special circumstances make the indigenous proportion much more important, the target value is supposed to identify the need to devise local/regional strategies complementing the EU-wide approach. In any case, the fact that the target value is to be attained "*as far as possible"* provides some flexibility when assessing compliance.

2.4. Consequences of a Target Value (TV) exceedance

2.4.1. Role of the Target Value

Based on the above considerations, the following consequences of TV exceedances for Member States can be identified and should be embodied in the daughter directive:

i) On the basis of a preliminary monitoring campaign and/or existing monitoring networks (possibly supplemented by modelling), Member States will identify the regions or zones where the TV is currently not met;

ii) Member States will be required to achieve the TV as far as possible throughout their territory within the required time period;

iii) Member States will be required to draw up plans demonstrating how they intend to achieve the TV within the required time period.

2.4.2. Linking the Target Value to relevant provisions of the Framework Directive (FWD)

To do this, the Commission and the Member States (MS) must have a clear picture of the frequency and occurrence of TV exceedances and of the measures taken or planned in the MS. This is expressed only once in Article 4(1)[9] of the Framework Directive (FWD).

It is proposed that the status of the TV and the associated assessment be embodied more clearly by referring to certain FWD provisions associated with the Limit Value (LV), namely Article 8(3)[10] and (4)[11], which require MS to prepare (integrated) abatement plans and programmes in the event of exceedances. In accordance with the definition of the TV in the FWD, an appropriate paragraph should reflect how transboundary pollution transport makes it difficult for MS to attain the ozone TV by individual measures alone.

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[9] See 2.2.3

[10] "... to ensure that a plan or programme is prepared or implemented attaining the LV within the specific time limit"

[11] "zones .... where the level of more than one pollutant is higher than the LV, MS shall provide an integrated plan covering all the pollutants concerned."

The plans referred to in Article 8(3) and (4) FWD, in which the MS outline their means of attaining the TV, will obviously be linked to, and need to take account of, the national plans which will be associated with the future proposal on national emission ceilings, and would in practice contain several actions based on sector-specific EU legislation. However, further elements in these programmes may consist of additional (including non-technical) measures implemented at MS level in order to achieve the national emission ceilings and/or to combat in the long term locally induced ozone levels above the TV.

To perform the evaluation required by Article 4(1) on the need for additional measures at EU-level, the Commission needs regular information on the zones in which TV exceedances occurred and on the MS's abatement plans. This should be embodied in the daughter directive by reference to Article 11(1)(a) and (b)[12] FWD, while a further link to Article 8(5)[13] FWD would specify in a concrete way the Commission's commitment to appraise the progress of the abatement strategy with regard to the ozone trend. According to the approach in the current Directive the Commission should also publish an annual list of zones with TV exceedances. In order to maintain the practice of preparing the annual and summer reports right after the summer season to be available in time for the October Council, the time period for Member States to deliver the relevant information should be shortened to six months.

The notion of Article 8(6) FWD, that joint abatement plans between Member States might be necessary to tackle TV exceedances originating mainly from emissions in another Member State, should be strengthened.

An overview of how the TV would be embodied according to the above considerations is presented in Fig. 2.1.

2.5. Consequences of a exceedance of a long-term objective (LTO)

The purpose of the LTO is to set a strategic reference point for ozone abatement policy based exclusively on effect-related considerations. Given the difficulty of attaining the LTO everywhere within a foreseeable period, the LTO should not be an immediate trigger for measures. However, reflecting the long-term goal of the 5th Environmental Action Programme to achieve the critical levels everywhere, it is proposed that the daughter directive embody a commitment for MS to aim at improvement of ozone levels even if the TV is already attained but the LTO is still exceeded. This would be consistent with the gap closure approach discussed within the strategy development, where efforts would not be restricted to areas where the highest pollution loads were found. When formulating a corresponding article in the Daughter Directive an appropriate distinction needs to be made *vis-à-vis* the stronger commitment linked to non-attainment of the TV.

In line with the FWD and the approach proposed for the TV, Member States are required to provide the Commission with the relevant information on the occurrence of LTO exceedances. However, it would be sufficient to request the information on potential measures for attaining the LTO only once, and to require no specific evaluation procedure. The Commission must publish a list of these zones annually.

<u>Relevance to Air Quality Assessment</u>

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[12] ".... inform the Commission of the occurrence of levels exceeding...., the reasons, the plans and programmes ...."

[13] "The Commission shall regularly check the implementation of the plans or programmes submitted ....., by examining their progress and the trends in air pollution"

As pointed out above, the LTO should be used as the trigger for the assessment obligation, i.e. the requirements regarding monitoring networks and supplementary assessment methods. For ozone the Target Value would represent a higher level than the long-term objective. If assessment were linked to the Target Value, inadequate assessment would be made of the extent to which the long-term objective is still exceeded. This irrationality is overcome if the long-term objective is used as the trigger for the assessment requirements.

This will be explained further in Chapter 4 of this paper, where it is proposed not to use the concept of upper and lower assessment levels.

2.6. The Alert Threshold and short-term action plans

2.6.1. Health-related information release in the FWD and the current Ozone Directive 92/72

The existing ozone Directive 92/72/EEC defines a *population information threshold* and a *population warning threshold* as follows:

> *"Information threshold means the ozone concentration value [...] beyond which there are limited, temporary effects on human health in the event of short exposure of particularly sensitive sections of the population and at which steps must be taken by the Member States as laid down in this Directive."*
>
> *"Population warning threshold means the ozone concentration value [...] beyond which there is a risk to human health in the event of short exposure and at which steps must be taken by the Member States as laid down in this Directive."*

These steps to be taken by the Member States comprise informing the population about exceedances and providing health-related advice.

The Framework Directive defines the *alert threshold* as:

> *"a level beyond which there is a risk to human health from brief exposure and at which immediate steps shall be taken by the Member States as laid down in the Directive."*

As in the current ozone Directive, the *necessary steps* (Article 10 FWD) to be taken by the Member States are to inform the population immediately of exceedances and give any precautionary advice.

So, the definition of the alert threshold and the consequences of an exceedance are almost identical to that of the warning threshold in the current ozone Directive. In principle the FWD does not necessarily require an alert threshold to be set. Nor does it refer to an *information value*. Nonetheless, in Section 3.7.4 it is proposed to retain the existing system of two threshold levels, indicating the requirement to issue health-related advice to sensitive groups and the general population, respectively.

Provision of this information (for details see chapter 6) is not intended as an alert or warning to the population, nor is the aim to link exceedance of either of these thresholds directly to short-term actions (see next section). However, alert thresholds were set for other pollutants (SO_2, NO_2) in the Common Position for the other daughter Directive. Rather than introduce a new type of threshold, with a function similar to the alert threshold, it is proposed to comply with the Framework Directive by basing the definition of such thresholds on the concept provided by that Directive. In reconciling the concept of two such levels with the provisions of the Framework Directive it is proposed to set an *information threshold* defined as an alert threshold for the sensitive population

and a *general alert threshold* which concerns everyone. Both will trigger immediate publication of health-related information (on the proposed levels, see section 3.7.4)

2.6.2. Short-term action plans

Article 7(3) FWD mentions short-term actions as a possible (supplementary) abatement approach, obliging Member States (MS) to:

> *"Draw up action plans indicating the measures to be taken in the short-term where there is a risk of the limit values and/or alert thresholds being exceeded, in order to reduce that risk and to limit the duration of such an occurrence. Such plans may, depending on the individual case, provide for measures to control and, where necessary, suspend activities, including motor-vehicle traffic, which contribute to the limit value being exceeded."*

Short-term measures may serve as a complementary approach to reducing ozone peaks, though their potential for reducing ozone will depend on the local natural circumstances and on the characteristics of the precursor sources and their reduction potentials (see Chapter 5.3 for details). This requires a flexible approach when establishing short-term actions in the Daughter Directive.

Article 7(3) FWD is therefore interpreted to mean that:

- short-term action plans should be drawn up, where an air quality problem (i.e. alert threshold exceedances) exists **and**

- where such short-term actions can be expected to contribute perceptibly to the aim of reducing the risk and limiting the duration of exceedances. Otherwise the purpose of measures "*to reduce that risk...*" would clearly not be met.

- the concrete design of short-term action plans (including trigger levels) "*depend on the individual case"*, i.e. this falls within the responsibility of Member States.

So, it is proposed here:

- to use exceedance of the general alert threshold as a trigger for the obligation to perform once a principal investigation of the ozone-reduction potential of short-term measures. Thus, short-term action plans would be generated and implemented on condition that evidence indicated a perceptible reduction potential for peak ozone values. While it is hardly possible to set universal criteria for the effectiveness of short-term measures, a reduction of peak ozone values by 10% is considered the minimum improvement to be aimed at.

- not to prescribe the design of the short-term action plans, including the levels triggering specific actions (speed limit, traffic bans, etc.).

- to provide in an annex to the directive criteria which should be taken into account when designing action plans in order to ensure a certain level of effectiveness (see Section 5.3).

Having explored the potential of short-term measures, Member States should be required to inform the Commission of the results of their investigations and of the decision whether short-term action plans seem appropriate to reduce the ozone load within episodes.

While such measures will mainly comprise durable and large-scale actions devised and implemented at EU level (see section 5), short-term actions may be also among such tools in those regions where the above investigations have shown an existing reduction potential.

The proposed approach is shown schematically in Fig. 2.2

Establishing target value and long-term objective

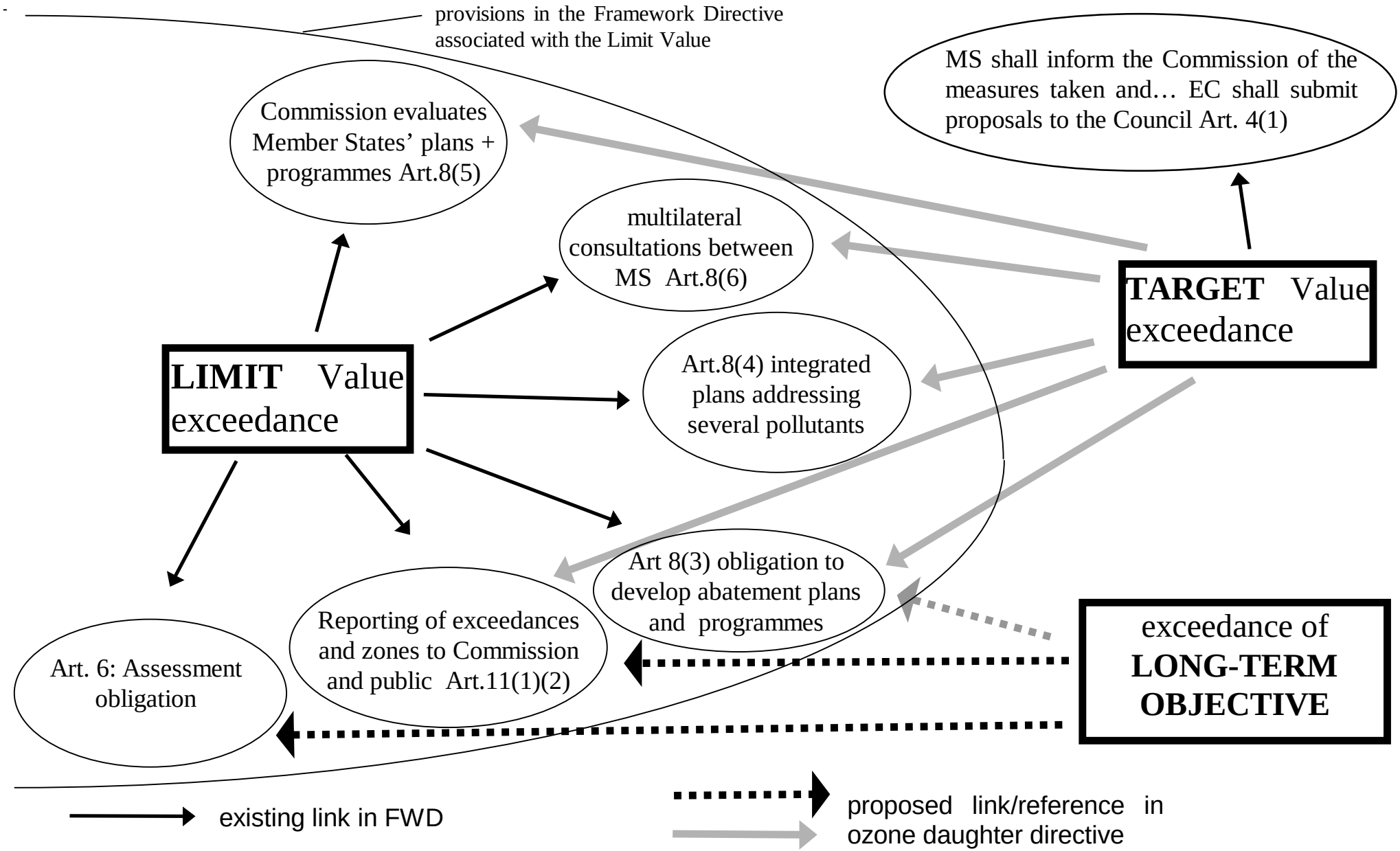

Fig. 2.1

Proposed approach in the daughter directive on short-term actions

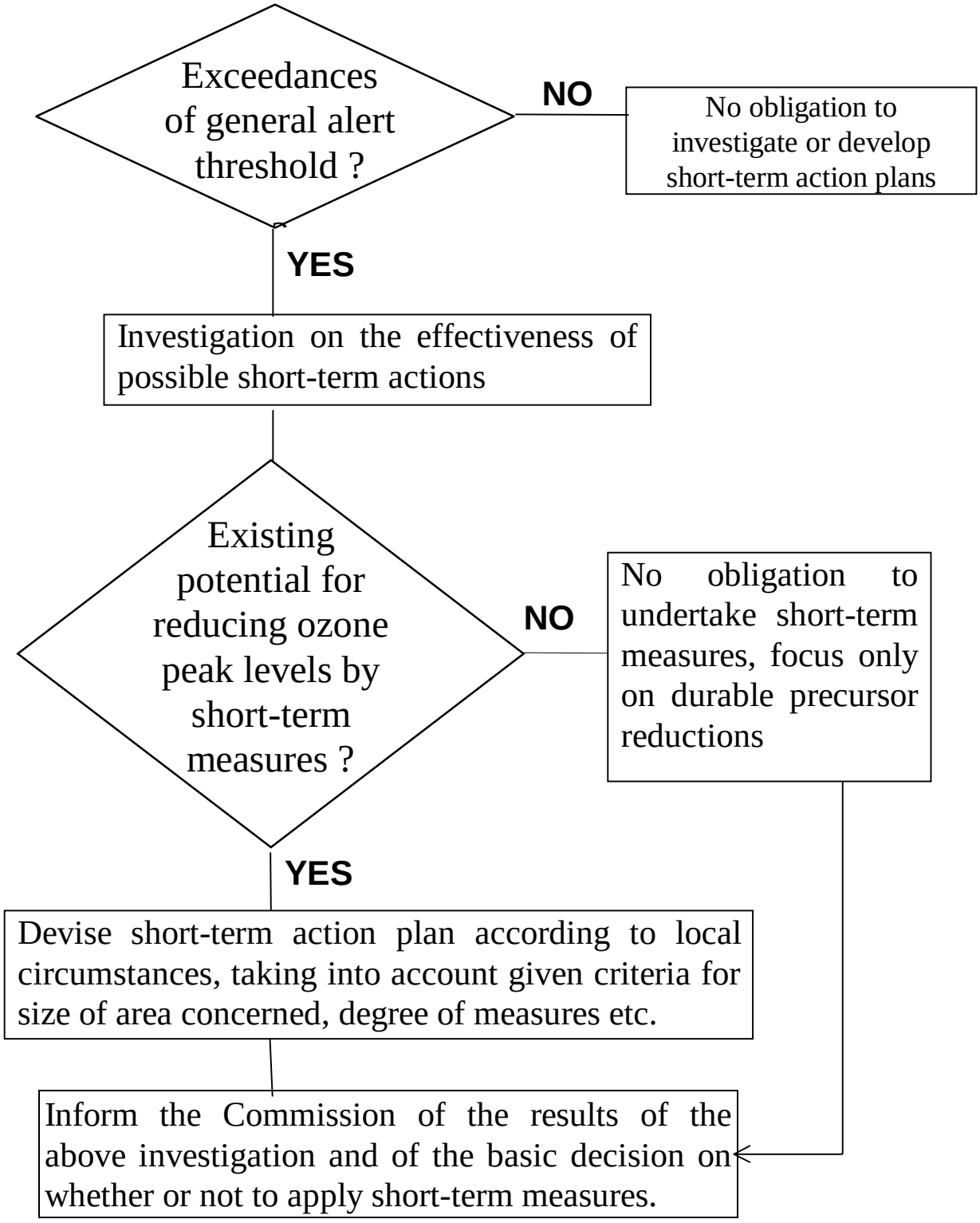

Fig. 2.2

REFERENCES

Council Directive 96/62/EC of 27 September 1996 on ambient air quality assessment and management; OJ L 296, 21.11.96, p.55.

Council Directive 92/72/EEC of 21 September 1992 on air pollution by ozone; OJ L 297.

COM (97) 88 final, OJ L 017, 21/01/1997, pp.17-27.

3. RISK ASSESSMENT

3.1. Risk from Ozone

In 1996 reviewed and updated its Air Quality Guidelines for Europe as published in 1987. This process included the evaluation of scientific knowledge on the effects of a large number of air pollutants, including ozone. Effects on human health as well as on ecological receptors were investigated. The current evaluation of risks from ozone is largely based on two WHO reports: "Ozone and other photochemical oxidants" (WHO, 1999a) and "Ozone effects on vegetation" (WHO, 1999b). The references given in the guideline text were not repeated in this chapter. Those appearing here refer mostly to recent studies which could not be considered by WHO.

3.1.1. Health effects

Ozone is a powerful oxidant, and as such can react with a wide range of cellular components and biological materials. In particular, damage can occur to all parts of the respiratory tract, the extent of which is dependent on the ozone concentration, exposure duration, exposure pattern, and ventilation. Effects observed in the respiratory tract include inflammation; morphological, biochemical, and functional changes; and decreases in host defence functions. In general, the effects after intermittent exposure appear to be more pronounced than following continuous exposure.

The time course of the changes in the respiratory system, as determined in laboratory animals as well as in epidemiological investigations, is complex. During the first few days of exposure, inflammation occurs and then persists at an attenuated level. At the same time, epithelial hyperplasia progresses, and reaches a plateau after about one week of exposure. After cessation of exposure these effects slowly disappear. In contrast to this, interstitial fibrosis increases slowly and can persist even after exposure ceases.

3.1.1.1. Effects on laboratory animals

Acute exposure of animal species for 2 to 3 hours to ozone concentrations of >400 $\mu g/m^3$ resulted in effects such as increased breathing rate, increased pulmonary resistance, decreased lung volume and vital capacity, and increased airway responsiveness. The latter effect is only induced at concentrations >2000 $\mu g/m^3$. These responses are generally resolved rapidly and disappear within 2 weeks after cessation of exposure.

Studies of exposure to ozone for several hours to a few days have shown alterations in the respiratory tract in which the lowest-observed-effect levels were in the range of 160-400 $\mu g/m^3$ (0.08-0.2 ppm). These included the potentiation of bacterial lung infections (160 $\mu g/m^3$), inflammation (240 $\mu g/m^3$), hyperplasia of Type II cells, increases in antioxidant enzyme activity and collagen content (400 $\mu g/m^3$). Long-term exposure to ozone in the range of 240-500 $\mu g/m^3$ (0.12 to 0.25 ppm) causes morphological changes in the epithelium and interstitium of the centriacinar region of the lung.

In addition to effects on the respiratory system, a wide range of extrapulmonary effects (e.g. decrease in activity level, changes in red blood cells and serum enzyme activities, morphological alterations on the thyroid and parathyroid) have been identified after exposure to ozone at concentrations of 240 $\mu g/m^3$ and higher. Whether these effects are caused by ozone itself or by reactive intermediates formed in the respiratory tract, or whether they are an expression of secondary reactions to pulmonary injury, is unknown.

Chronic exposure to a high concentration of ozone (2 mg/m^3) has been shown to evoke a limited degree of carcinogenicity in females of one strain of mice. Rats were not affected. Furthermore

there was no concentration dependent response, and there is inadequate information from other research to provide mechanistic support for the finding in mice. Thus the potential of ozone for animal carcinogenicity is uncertain. Abbey *et al.* (1991) have suggested an increase in respiratory cancer for chronic exposure to relatively high concentrations of a/o. total suspended particulate matter and ozone. This association cannot be disentangled from exposure to TSP and ozone. It is for that reason not possible to evaluate the effect specifically related to ozone. No other observational studies are available to evaluate the role of chronic ozone exposure in inducing pulmonary carcinomas or other malignant neoplasms. It is the general judgement of health experts that no reliable evidence is available that ozone is carcinogenic in man (US-EPAa).

In general, the mechanistic findings in these animal studies support the observation in humans.

3.1.1.2. Effects on humans

In a large number of controlled human studies, significant impairment of pulmonary function has been reported. Studies with acute, single ozone exposure have lasted from 1 to 8 hours with exposure concentrations ranging from 160 to 1000 $\mu g/m^3$. Exposure to ozone of normal subjects for 1 to 3 hours during moderate-to-heavy exercise caused the following changes: decrease in forced expiratory volume (FEV_1) at $\geq$240 $\mu g/m^3$, increase in airway resistance at $\geq$360 $\mu g/m^3$, decreased forced vital capacity (FVC) at $\geq$240 $\mu g/m^3$, and increased respiratory frequency ($\geq$400 $\mu g/m^3$). With 4 to 8 hours of ozone exposure, healthy young adults engaged in moderate exercise showed changes in the following pulmonary function tests: decreased FEV_1 ($\geq$160 $\mu g/m^3$), increase in airway resistance ($\geq$160 $\mu g/m^3$), decreased FVC ($\geq$200 $\mu g/m^3$), increased airway responsiveness ($\geq$160 $\mu g/m^3$). In summary it can be said that these health effects are statistically significant at 160 $\mu g/m^3$ (0.08 ppm) for 6.6 hour exposures in a group of young healthy exercising adults, with the most sensitive subjects experiencing >10% functional decrements within 4 to 5 hours.

An open discussion has taken place on contamination of ozone used in the human chamber studies, but it was concluded that possible interference's were too small to affect the determination of exposure-response relationships for ozone.

Field studies in children, adolescents, and young adults have indicated that pulmonary function decrements, similar to those observed in controlled studies, can occur as a result of short term exposure to ozone concentrations in the range of 120-240 $\mu g/m^3$ and higher. Mobile laboratory studies using ambient air containing ozone have observed associations between changes in pulmonary function in children or asthmatics and ozone concentrations of 280-340 $\mu g/m^3$ (0.14-0.17 ppm) with exposures lasting several hours.

Children represent for several reasons a very sensitive group of the population (Budinger, 1996). In comparison with adults, children have a higher intake of ozone and other air pollutants. This is due to a higher basal metabolic rate, resulting in a higher breath volume per minute and a higher breathing frequency. Further, their respiratory tract is still under development until the age of six and a half, and is therefore more susceptible to the inflammatory effects of ozone. Children's immune systems are not yet fully developed either and are generally under bigger stress. For these and other reasons, children are at higher risk concerning exposure to ambient ozone concentrations.

Respiratory symptoms, especially coughs, have been associated with ozone concentrations as low as 300 $\mu g/m^3$ (0.15 ppm). Ozone exposure has also been reported to be associated with increased hospital admissions for respiratory causes and exacerbation of asthma. That these effects are observed both with exposures to ambient ozone (and co-pollutants) and with controlled exposures to ozone alone demonstrates that the functional and symptomatic responses can be attributed primarily to ozone.

Patients with lung disease and smokers are thought to be at increased risk of ozone-induced decrements in lung function because an equivalent decrement in lung function would have more serious health consequences given their already compromised lung function.

In contrast to healthy subjects, children with moderate to severe asthma had greater dose-related changes in lung function and these responses were occurring at relatively low ambient concentrations of O_3. The highest peak daily O_3 concentration was 300 $\mu g/m^3$, but the responses were still statistically significant when excluding days with peak concentrations above 180 $\mu g/m^3$.

Other groups at risk are, particularly, people exercising outdoors during the time of day that ozone concentrations tend to be highest. These groups comprise children, manual workers and athletes, who might have an increased ozone dose rate as a result of their increased inhalation rate.

There is limited information linking long-term ozone exposure to chronic health effects. While there is a suggestion that cumulative ozone exposures may be linked to increasing asthma severity and the possibility of increased risk of new asthma, this association is not clearly established. This indicates the need for further research into the chronic effects of ozone exposure in order to take this aspect into consideration in the next guideline review process.

The association of daily changes in mortality with changes in ozone levels has also been studied. Until recently the limited number and inconsistent outcomes of studies on all-cause mortality did not provide strong evidence that ozone is causally related to mortality. However, the recent analysis based on the APHEA study, including data from four other European cities, provided evidence that daily mortality increases by 2.3% (1.4-3.3%) per 50 $\mu g/m^3$ increase in the maximum daily 1-hour mean of ozone (Touloumi *et al.*, 1997).

3.1.1.3. Ozone and asthma

Both healthy people and individuals suffering from respiratory diseases vary greatly in their response to ozone and oxidant air pollution (McDonnell, 1991). A distribution has been suggested including non-responders, normal responders, and hyperresponders. This large phenotypic variation may depend on inhaled doses in the airways and biological sensitivity determined by intrinsic factors, e.g. genetic background and disease status, and extrinsic factors, e.g. socio-economic status (Tankersley and Kleeberger, 1994). In addition, the variation between individuals can be different for the various effect endpoints, i.e. a hyperresponder with regard to e.g. decline of FEV_1 might be a non- or normal responder for an inflammatory reaction and vice versa. Standard-setting for ozone is intended to protect the majority of the sensitive subgroups, and quantification of ozone risks has taken this response variation into account by also predicting the proportion of the most sensitive sub-populations in the general population, based on exposure and exposure-effect models.

Asthma patients are considered an important risk population because some 5-10% of the general population suffer from asthma. Asthma encompasses clinical conditions that include airflow limitation (obstruction) and (chronic) inflammation of the airways. Due to the irritant nature of ozone, capable of inducing airway inflammation and bronchoconstriction, asthma patients are deemed to be at enhanced risk from exposure to ozone and photochemical ‘smog’, because inflamed airways contribute to the pathogenesis and exacerbation of the disease and to morbidity and mortality for asthma.

Epidemiological studies indicate that exposure to ozone during oxidant (summertime) air pollution is associated with exacerbation of asthma, increased hospital admissions and visits to emergency departments for asthma attacks, and increased medicine use by asthmatics (Cody *et al.*, 1992; White *et al.*, 1994). In addition, urban ozone levels appear to be associated with nasal epithelial changes and nasal inflammatory reactions in healthy young males (Calderon-Garcidueñas and Roy-Ocotla, 1993) and children (Frischer *et al.*, 1993). Recent field studies indicate that asthmatic

children and adults exposed all day to ozone levels at or below national standards experience a degree of effects similar to those occurring after exposure to higher concentrations for one or two hours, as observed in controlled human studies (Thurston *et al.*, 1997). Epidemiological studies have not indicated an apparent threshold value for ozone, and effects seem to be linear with ozone concentrations in ambient air.

Controlled human studies show that relatively low concentrations of ozone cause increases in airway hypersensitivity and maximal degree of airway obstruction in asthmatics (Seltzer *et al.*, 1986; Hiltermann *et al.*, 1995). Studies also show that ozone concentrations which do not reduce pulmonary function or reduce it only slightly cause greater airway inflammation in asthmatics than in healthy subjects (Aris *et al.*, 1993; Basha *et al.*, 1994; McBride *et al.*, 1994; Scannell *et al.*, 1996) In addition, recent studies also indicated that short-term ozone exposure increased the bronchial responsiveness of subjects with allergic asthma to an allergen challenge and that the total ozone dose was considerably lower than the dose usually applied to elicit lung function decrements (Seltzer *et al.*, 1986; Molfino *et al.*, 1991, 1992; Folinsbee *et al.*, 1994; Jörres *et al.*, 1996).

Collectively, these data indicate that asthmatic patients are at increased risk from ozone exposure due to airway inflammation, airway reactivity and obstruction, as well as a stronger than usual reaction to inhaled allergens. In addition, even if asthmatics present a degree of respiratory symptoms and lung function decline similar to those displayed by healthy people their lung condition is still being further compromised.

3.1.1.4. Health impact considerations

The photochemical ambient oxidant mixture consists of a variety of different compounds and although ozone is considered the most important cause of adverse effects in the airways, other compounds may very well contribute to these effects. Ozone levels are used as an index to indicate photochemical air quality and to assess the severity of effects (WHO, 1992).

Current scientific evidence of health effects caused by exposure to ozone and photochemical air pollution suggests that a WHO Air Quality Guideline of an 8-h maximum value of 120 µg ozone per m^3 is a level at which acute effects in the population are present. Asthma patients are especially at increased risk due to inflammation, worsening of asthma, and hospital visits. The database has not revealed a threshold value for ozone.

Both epidemiological and controlled human data can be used to establish estimates of health effects in relation to ozone levels, as depicted in Tables 3.1 and 3.2 in section 3.2.

An example of such an approach was given in the Consolidated Report (Beck *et al.*, 1999), where reported exceedances of the threshold for the protection of human health in the current Ozone Directive 92/72/EEC were used to estimate the population exposure to ozone. Based on the estimated exposure distribution and relative risk estimates from the APHEA study, it can be calculated that 0.1-0.3% of hospital admissions could be attributed to ozone concentrations exceeding 110 µg/m^3. This corresponds to 300-1000 hospital admissions for respiratory conditions in the entire population of all 15 EU countries.

Broadly, the relative risk of respiratory admissions for Europe is similar to that reported for the USA. In addition, in neither continent was there significant heterogeneity between the cities involved in the studies, which provides reassurance regarding the generalisation of the data.

Based on the results of the above mentioned studies on exposure-response associations (3.1.1.2) and using estimates of population exposure based on reporting of exceedances of the EU threshold level (Beck *et al.*, 1999), it can be estimated that approximately 1 500-3 700 advanced deaths could have been caused in the 15 EU countries by ozone episodes in excess of 110 µg/m^3 in the summer of 1995. This is about 0.1-0.2% of all deaths. It is likely that the total number of

advanced deaths is several times higher than the estimate for days with high ozone levels (>110 $\mu g/m^3$) only.

A similar approach has been used in the UK, mapping ozone concentrations across the UK each day for summer periods, taking into account corrections in the urban concentration fields, combined with exposure-response relationships for health outcomes derived from epidemiological studies in the UK. The Committee on the Medical Effects of Air Pollutants, advising the UK Department of Health, published the results of this study in January 1998 (COMEAP, 1998) in which they estimated that 700-12 500 advanced deaths and 500-9 900 hospital admissions were associated with ozone concentrations. The ranges are largely due to uncertainty over the assumption of a threshold for effects. Thus the lower end of the range of effects assumes a threshold of 50 ppb and the upper end of the range assumes no threshold.

It should be noted, however, that the calculations mentioned above are estimates based on limited data and a number of assumptions. The uncertainties in the outcome are reflected in the ranges provided. These estimates should therefore be handled with care.

3.1.2. Risks to vegetation

The potential for ozone damage to vegetation has been known for over thirty years, but it is only over the last decade that its impacts have become of concern in Europe. It is now clearly established that ozone at ambient concentrations found in Europe can cause a range of effects including visible leaf injury, growth and yield reductions, and altered sensitivity to biotic and abiotic stresses. Furthermore, because ozone is a secondary pollutant with a regional distribution, these effects may occur over large areas of rural Europe. Research in recent years has advanced our understanding of the mechanisms underlying ozone effects on agricultural crops, and to a lesser extent on trees and native plant species. It is now possible to determine biologically meaningful, but simple indices to characterise ozone exposure, and to identify the critical levels of exposure above which - by definition – direct adverse effects on receptors, such as certain plant species, may occur. Exposure to elevated ozone concentrations causes effects on individual crop and tree species, and on natural vegetation species mixtures, leading to losses in economic value, quality traits, and biodiversity. Negative effects on crop yield cause economic losses in agriculture which may be significant, while not necessarily inducing visible injury to leaves. However, for crops which are sold for their foliage (e.g. spinach and tobacco, which are both very sensitive to ozone), visible injury, even if it does not result in significant biomass reductions, may also cause important economic losses. Injury such as chlorosis and necrosis can reduce the market value of the crop. Long-term effects on trees may impair the function of forest ecosystems, i.e. their role with respect to the water and energy balance, protection of soil from erosion, etc. Important impacts on plant communities may not be through growth or productivity, or through visible injury, but through shifts in species composition, loss of biodiversity, and changes in genetic composition. While the evidence for effects on natural vegetation is limited, the establishment of a critical level is consistent with the precautionary approach and the resolutions to preserve biodiversity adopted at the Rio summit in 1992.

Ozone enters plants through the stomata pores on the leaf surface. Once inside the leaf, it rapidly reacts with the moisture on the surfaces of the underlying cells. The resulting oxygen species are highly reactive and may overload the naturally occurring protective mechanisms of the plant. Cell membranes may be damaged, leading to the destruction of cells, and ultimately to the appearance of necrotic or chlorotic areas on the leaf surface. Such injury is usually a response to ozone episodes of 2-3 days' duration. If the ozone stress is prolonged, physiological changes occur such as reduced photosynthesis, which can lead to a reduction in yield or an alteration in the allocation of biomass between the leaves, roots and seeds.

Ozone episodes often occur throughout the growing period of crops and natural vegetation. The cumulative effect of damage to cell structure and reduction in key processes such as photosynthesis is manifested as a reduction in seed yield at the end of the growing period. Typically, ozone leads to a

reduction in the maximum leaf area of plants and a shorter duration of leaf lifetime. Leaves are frequently lost from the plant relatively early, resulting in an overall reduction in the amount of light intercepted by the plant. As the fruit develops, the consequent reduction in fixed CO_2 means that the plant is unable to provide sufficient assimilate to support the development of all seeds. Pods or ears may be shed from the plant, and individual seeds may be aborted. Yield reduction due to ozone is usually due to a decrease in the number of seeds per ear or pod and a decrease in individual seed weight.

For trees, experimental data are scarce. In Europe, two exposure-response studies of potted tree seedlings exposed to ozone in field fumigation chambers have been published. One of them focused on the effect on biomass in birch, the other on photosynthesis in Norway spruce. In Denmark, exposure-response data have been obtained for Norway spruce exposed to ozone in branch chambers. The effect of non-filtered versus filtered air has been tested on seedlings of beech and Norway spruce at different altitudes in Switzerland with prevailing ozone air pollution. Beech reacted to ambient ozone levels with a 10% biomass reduction over three years. It should be noted, however, that considerable difficulties exist in scaling-up from seedlings to mature trees. Consequently, the available exposure-response data from studies with seedlings may not reflect the response to ozone of older, mature trees or forest stands, and effects from long-term exposure on trees with a long life-cycle are unknown. At this stage only data for seedlings are available to define provisional critical levels for forest trees.

In order to relate ozone exposure to effects, it is necessary to summarise concentrations averaged over 1-hr intervals in a biologically meaningful way which can serve as a surrogate for dose. In principle, the exposure index must be based on the concept of effective dose, i.e. it must capture the characteristics of exposure which most directly relate to the amount of ozone that is absorbed by vegetation. Uptake of ozone could be estimated by multiplying the concentration near the leaf surface by the leaf conductance for ozone, and the absorbed dose would then be the integral of the rate of uptake (flux) over time. This concept could be expanded to take into account the conductivity of the atmosphere. In situations with sufficient air mixing (high air conductivity), the diurnal pattern of ozone flux is determined by leaf conductance and ozone concentration; this is the case in open-top exposure chambers. Due to the lack of leaf conductance data, the use of radiation as a surrogate for leaf conductance has been suggested in agricultural crops, and the most simple approach is to use ozone concentrations measured during daylight hours (e.g. >50 W m^{-2} global radiation) to characterise exposure. For species with substantial leaf conductance at night, however, no such discrimination should be made; available data on leaf conductance are limited to a few species at present. Other factors, e.g. air humidity, soil water availability, or temperature, are also known to influence leaf conductance, but to date these factors have not been used to characterise ozone uptake or dose in long-term experiments.

Long-term exposure to ozone can lead to growth and yield reduction. Hence the most suitable exposure indices to be related to long-term effects are cumulative, i.e. they integrate exposure over time. Previously, air quality guidelines for long-term effects have been based on mean concentrations over a given period of time, e.g. the arithmetic mean over the growing season of the daily mean concentrations during a specific 7-h period (usually 09.00-16.00). The use of a mean calculated for a given period of time implicitly gives equal weight to all concentrations. However, experimental exposure-response studies with ozone suggest that this is not appropriate, and that it is the intermittent exposure to higher concentrations which is most important in causing long-term effects. This can be explained physiologically by the capacity of the plant to detoxify ozone and other oxidants; it is only when the concentration, or flux, of ozone exceeds this capacity that adverse effects result. Hence in attempting to assess the adverse effects of exposure to ozone, it is essential to consider both the duration and the extent of exceedance of any guideline value, not just the total exceedance.

3.1.3. Effects on materials

Ozone can act in different ways in the degradation of materials. The direct effect on organic materials is relatively easy to assess. However, ozone is part of a multi-pollutant situation, which

means that direct effects only account for part of the total influence, which also includes indirect and synergistic effects.

3.1.3.1. Direct effects

While degradation of inorganic materials is mostly associated with SO_2 and NO_2, the degradation of organic materials has traditionally been associated only with O_3 in addition to the most important natural factors, temperature and solar radiation (Tidblad J. and V. Kucera, 1996). The major effects of O_3 have been noted in organic polymers possessing double bonds in their structure. Two damage mechanisms may occur: chain-scissoring results in a reduction in average molecular weight and loss of tensile strength, while cross-linking of polymers increases rigidity and reduces elasticity, and may result in brittleness (Lanting, R. W., 1984). O_3 is a principal pollutant primarily associated with the degradation of rubber, though most organic material, such as painted surfaces, polymers and textiles, is sensitive to it. Exposure to O_3 leads to fading and embrittlement of paints, cracking of rubber and fading of dyes in textiles or reduction of textile strength (Lee, D.S et al., (1996), Lewry A., 1991). The affected materials may be subdivided into thin organic layers such as paints and coatings, bulk organic items such as gaskets, sealants or PVC windows, and rubber materials such as car windscreen wipers or rubber roofing materials. However, its effect can be difficult to distinguish from direct sunlight damage (Yocom, J. E. and McCaldin, R. O., 1968). At the concentrations found in the indoor atmosphere of many museums, O_3 poses a risk of fading of the pigments used in works of art. The pigments have different sensitivities depending on the base material (Grosjean D et al., 1993).

For inorganic materials O_3 is primarily regarded as an oxidiser of SO_2, but possibly has other roles such as promoting oxide formation and affecting the protective abilities of anti-corrosion products (Tidblad J. and V. Kucera, 1996).

3.1.3.2. Multi-pollutant effects

O_3 is a general oxidant. It is involved in atmospheric and corrosion processes, and therefore has a dominant role in the multi-pollutant situation. Due to diurnal and seasonal variations the correlations between pollutants are high. This is especially true for O_3 and NO_2, which are often negatively correlated, if annual averages are used. Since both NO_2 and O_3 can have similar effects on various materials, a statistical analysis of field exposure data will always have difficulty separating their individual effects (Tidblad J. and V. Kucera, 1996).

a) Indirect effects

The presence of O_3 in the atmosphere may result in secondary pollutants. The reaction of O_3 and NO_2 may result in HNO_3, both indoors (Weschler, C. J., et al., 1992a and 1992b) and outdoors. This is a pollutant with high potential for damage to a wide range of materials. Once formed, HNO_3 is extremely reactive and will readily deposit on virtually any contact surface (Lipfert, F.W., 1987).

b) Synergistic effects

Synergistic effects seem to be present for most inorganic materials. In practice, the term "synergistic effects" refers almost exclusively to the combination of SO_2 with a second pollutant, often NO_2 or O_3. The effect of the second pollutant has been attributed to the oxidation of S(IV) to S(VI), i.e., sulphite to sulphate. Very few studies have investigated the combined effects of SO_2 and O_3. When comparison is possible, the synergistic effect of SO_2 in combination with O_3 is often stronger than the synergistic effect of SO_2 in combination with NO_2. Little is known about other explanations for synergistic effects, besides SO_2 oxidation (Tidblad J. and V. Kucera, 1996).

3.2. WHO recommendations

3.2.1. Health guideline

This section describes the derivation of a WHO Health Guideline, taken mainly from the relevant chapter of WHO (1999a). Establishing a guideline value for ozone in ambient air is complicated by the fact that detectable responses occur at or close to the upper bounds of background concentrations. Therefore it is not possible to base a guideline value on a NOAEL or LOAEL with an uncertainty factor of more than a small percentage. In controlled human studies there are statistically significant decrements in lung function, airway inflammatory changes, exacerbations of respiratory symptoms and symptomatic and functional exacerbations of asthma in exercising susceptible people, as well as increased hospital admissions for respiratory causes at ozone levels of 160-360 µg/m3 (for 1 to 8 hours exposure periods); where 160 µg/m3 is the lowest level tested. Field studies indicate that some pulmonary function decrements might be found at ozone concentrations possibly as low as 120 µg/m3. In order to select a guideline value, one must accept the premise that some detectable functional responses are of little or no health concern, and that the number of responders to effects of concern are too few to represent a group warranting protection from exposures to ambient O3. In the case of respiratory function responses, a judgement could be made that O3-related reductions of FEV1 of less than 10% were of no clinical concern. As mentioned above (3.1.1.2) significant effects of 10% or more occurred at ozone levels of 160 µg/m3 and higher. The level of no clinical concern should therefore be below this concentration.

Based on these assumptions and an overall judgement of the experimental observations, WHO established a guideline value for ozone in ambient air of 120 µg/m3 for a period of 8 hours per day as a level at which acute effects on public health are likely to be small. Although chronic exposure to ozone has the capability to cause effects, quantitative information from humans is considered inadequate yet to estimate the degree of protection from chronic effects offered by this guideline.

Previous recommendations have included a 1-hour guideline value of 150-200 µg/m3, and an 8-hour guideline of 100-120 µg/m3. These recommendations did incorporate a margin of safety of 50 percent, and were based upon the lowest observed adverse health effects available at that time. The present 8-hour air quality guideline of 120 µg/m3 does not include such a margin of safety and is actually based upon an acceptance of a certain amount of risk to the general population. Although recent research does not indicate that the 1-hour guideline would necessarily be erroneous, it was concluded by WHO that the 8-hour guideline would protect against acute 1-hour exposures in this range and that a 1-hour guideline would not be necessary.

Table 3.1

Health outcomes associated with controlled ozone exposures		O3 concentration (µg/m3) at which health effect expected.	
	Averaging time	1 h O3	8 h O3
FEV1 change (active, healthy, outdoors, most sensitive 10% of young adults and children)			
5%		250	120
10%		350	160
20%		500	240
Inflammatory changes (influx of neutrophilic cells) (healthy young adults at >40 l/min outdoors)			
2-fold increase		400	180
4-fold increase		600	250
8-fold increase		800	320

Based on expert judgement of all the collective evidence from numerous controlled exposure studies and the assumption of a linear relationship between exposure and effects, WHO estimated the association between two representative health outcomes and ozone exposure.

The results of this exercise are presented in Table 3.1. It should be recognised, however, that these associations are estimates and that the ozone concentration at which any adverse health outcome is expected will vary with the duration of the exposure and the volume of air that is inhaled during the exposure (level of exercise).

From epidemiological data WHO also established relationships between changes in health outcomes and changes in the peak daily ambient ozone concentration. Recent studies, including the APHEA study analysing data from five large European cities with respect to the association between hospital admissions and ozone levels, point to a somewhat lower risk than previous estimates. From the meta-analysis of the data of the APHEA study, the increase in frequency of hospital admissions associated with ozone exposure was calculated: RR (relative risk) = 1.043 per 50 µg/m3 of 8-hour mean ozone levels. This risk estimate generally agrees with observations from North American studies.

A summary of health outcomes associated with changes in ambient ozone concentrations is shown in Table 3.2. Short term increases in levels of ambient ozone are associated both with increased hospital admissions with a respiratory diagnosis and respiratory symptom exacerbations in both healthy people and asthmatic patients.

Table 3.2

Health outcomes associated with changes in ambient ozone concentration in epidemiological studies	Change in 1h O3 (µg/m3)	Change in 8h O3 (µg/m3)
Symptom exacerbations among adults or asthmatics - normal activity		
25% increase	200	100
50% increase	400	200
100% increase	800	300
Hospital admissions for respiratory conditions*		
5% increase	60	50
10% increase	120	100
20% increase	240	200

* Given the high degree of correlation between the 1-hour and 8-hour ozone concentration in field studies, a reduction in health risk associated with decreasing 1h or 8h ozone levels should be almost identical.

The exposure-response relations given above may be used to quantify expected improvements in health outcomes that may be associated with lowering the ambient ozone concentration. It should, however, been kept in mind that these results are based on linear relationships and that it is uncertain whether similar response slopes can be expected at widely different ambient ozone levels.

3.2.2. Ecosystem guidelines

WHO recommends a number of guideline values for ozone (WHO, 1999b). These are based upon the Critical Levels for ozone developed under the UN/ECE Convention on Long Range Transboundary Air Pollution. LRTAP Convention workshops have defined Critical Levels, and WHO workshops have defined the Air Quality Guidelines. Both Critical Levels and WHO Guidelines are based upon the same scientific information and experimental results, so there is consistency between the agreed values for Critical Levels and Guidelines. The Guideline values are highlighted in bold italics in the text and summarised in Table 3.3 below.

For the work under the LRTAP Convention a Critical Level is defined as "the concentration of pollutants in the atmosphere above which direct adverse effects on receptors, such as human being, plants, ecosystems or materials, may occur, according to present knowledge". In order to determine Critical Levels, quantitative relationships between the pollutant exposure and the effect of interest are needed. However, any such relationships have a certain degree of uncertainty, and the data necessary to produce them are often scarce. A consensus was reached in the workshops as to how exposure should be expressed, and what type of effect, and what acceptable level of effect, should be used as the criteria to define the Critical Level. Based on the results from recent experiments with a limited number of agricultural crop species, the cumulative exposure index using a threshold of 40 ppb has been accepted as the best available exposure index, using hourly concentrations during daylight hours over a 3-month period (the growing season). This concept was adopted at the UN/ECE workshop at Egham in 1992, when a threshold concentration of 40 ppb was tentatively suggested. This exposure index has been called the AOT40, i.e. accumulated exposure above a threshold concentration of 40 ppb, expressed in units of ppb. hours or ppm. hours. Statistical analysis of yield data from European open-top chamber experiments has demonstrated that the use of this threshold generally provides better linear fits to exposure-response data than the use of higher thresholds. A linear exposure-response relationship provides a sounder statistical basis than other types of relationships for defining a Critical Level corresponding to a specific effect. The use of 40 ppb as the threshold has been favoured over lower

threshold concentrations because in Europe this value broadly corresponds to the boundary between mean concentrations in areas with low and high frequencies of photochemical episodes. However, the choice of this threshold does not imply that concentrations below 40 ppb have no effect. Hence, the threshold concentration does not present a threshold for effects, but rather a cut-off concentration. Because of the increase in the proportion of background ozone with increasing altitude, the use of a cut-off concentration of 40 ppb may not be appropriate for higher elevations. Plant adaptation at such altitudes may raise a further complicating factor.

The exposure-response relationship between AOT40 and the grain yield of spring wheat can be fitted by a linear model. No threshold level of exposure was found to exist, and a Critical Level corresponds to any acceptable level of yield reduction. Hence, the Critical Level could be defined as the AOT40 corresponding to a yield reduction of 5% or 10%. The corresponding AOT40 values are 3 ppm. hours or 6 ppm. hours respectively (rounded to the nearest ppm. hour). Statistical analysis showed that the least significant deviation from 100% yield which can be estimated with 99% confidence is 4-5% . Therefore, it is recommended to evaluate the risk by calculating the exceedance of the Critical Level of agricultural crops, of an AOT40 of 3 ppm. hours per year (for the growing season and daylight hours).

A short term critical level for ozone can also be defined for the development of visible injury to crops. Results from UN/ECE ICP-Crops up to 1995 have indicated that species such as bean, clover, tomato and Soya bean develop foliar injury in response to ozone episodes occurring over 5 days. Development of visible injury is of particular importance for crops whose market value depends upon their appearance, e.g. spinach, tobacco. Furthermore, ozone-induced injury is an indication of the occurrence of elevated ozone concentrations.

Using the AOT40 index calculated during daylight hours, based upon the ICP crops analysis of data from white and subterranean clover, two critical levels for injury development have been defined in the WHO guidelines. These are:

- 500 ppb.hours over 5 days when mean vapour pressure deficit (9.30-16.30) exceeds 1.5kPa
- 200 ppb.hours over 5 days when mean vapour pressure deficit (9.30-16.30) is below 1.5kPa

As for the long-term critical level, these short-term critical levels refer to daylight hours only and should not be applied when soil moisture is limited.

However, data from experiments carried out in recent years suggest a less clear threshold for the effects of water vapour deficit. Further analysis of these data is in progress to identify critical levels across the range of water vapour deficit.

As in the case of crops during the work under the LRTAP Convention, it was agreed that exposure to trees be characterised by the index AOT40. For trees this should be calculated for daylight hours over a 6-month period covering the period of highest sensitivity of the tree. Under mid-European conditions, the six-month period would start at the beginning of April; however, for some Mediterranean species the period from April to October would not be appropriate. For protection of European forests, the critical level for ozone was proposed earlier to be an AOT40 value of 10 ppm. hours using all hours of the day. Only data for plant biomass obtained in the field or in open-top chambers were considered, whereas other parameters, such as chloroplast size or photosynthetic rate, were not regarded as suitable criteria. Also, results from studies in which other pollutants (e.g. SO2 or NO2) were added at concentrations close to their critical level are difficult to interpret. Last but not least, considerable uncertainties still exist with respect to both the response of mature trees and the long-term effects of ozone exposure. Reanalysis of the data for beech, representing the most sensitive receptor, yielded a significant regression of biomass decrease versus AOT40 for daylight hours. For this analysis, the data from experiments of

different duration were normalised to one growing season. The parameters obtained after normalisation provided an estimated critical level of 10 ppm. hours, which was associated with an 10% decrease in biomass. Hence, an AOT40 of 10 ppm. hours for daylight hours, accumulated over a 6-month growing season, was proposed as a critical level to protect forest trees.

Since there are very limited exposure-response data available for herbaceous plant communities, and these tend to relate to relatively short-term experiments on artificial mixtures of plant species, it is not possible to derive critical level values directly. However, there is evidence of significant changes in species composition at values of AOT40 similar to the critical levels set for agricultural crops. As in the case of crops, it is possible to calculate from the exposure-response relationship the ozone exposure corresponding to a given effect. Selecting a reduction by 10% in the proportion by biomass of the sensitive species as the criterion, the equivalent ozone exposure was 6.3 ppm. hours for Swiss data, and 5.7 ppm. hours for UK data on cut swards; a subsequent UK experiment using calcareous grassland rather than acidic grassland species gave a value of 7.8 ppm. hours. All these values are very similar, and are also comparable to the value of 6 ppm. hours for a 10% loss in yield in wheat. A more recent analysis of data from experiments on species of semi-natural plant communities indicated that reductions in shoot biomass or seed production of individual plants may occur when an AOT40 value exceeds approximately 3 ppm. hours. However, it is not known what the cumulative effect of these ozone exposures on species composition would be if they continued for several years. Based on the limited data available, it was suggested that ***the critical level for herbaceous plant communities (natural vegetation) should be no higher than that set for crops corresponding to a 5% yield loss, i.e. an AOT40 of 3 ppm. hours.***

When determining whether ozone exposures at a specific location exceed these critical levels, one has to take into account that AOT40 values can vary substantially from year to year because of the variability of the weather. Furthermore the critical level for crop yield was based an analysis of data in several different growing seasons, since a change in yield loss of e.g. 5% is in most instances not detectable on a single year's basis. Also the critical level for forests was based on multi-year experiments. It is therefore recommended that the exceedance of these critical levels, and of that for semi-natural vegetation, be evaluated on the basis of mean AOT40 values over a five-year period. However, where visible injury to crops resulting from short-term exposures is of direct economic concern, examination of monitoring data for the year with the highest ozone exposures is recommended.

The data used to derive critical levels are almost entirely drawn from experiments in open-top chambers in central and northern Europe, using plants that are adequately supplied with water and nutrients. There are uncertainties in using these data to define air quality guidelines for vegetation throughout Europe. Among the most important of these are:

1. The open-top chamber may alter the uptake of ozone to some extent, because of different air movements, humidity and light inside the chambers. Over- and under-estimations of effects in the experiments can thus not be excluded.

2. Many species have not been investigated experimentally in Europe, especially those of the Mediterranean region.

3. The critical level is likely to be higher when water supply is limited, because ozone flux is reduced, e.g. stomata closure in drought conditions. This is important in many areas of Europe, especially as periods of water stress often coincide with periods of high ozone concentrations. However, the irrigation of crops in dry conditions will complicate these predictions.

4. The data for trees are more variable than those for annual crops, and there is uncertainty in extrapolating from experiments of limited duration with young pot-grown trees to long-term effects on forest ecosystems. However, a recent field study of mature beech trees in Switzerland has observed

growth reduction in the range of 6-13% for exposure to AOT40 values of 10 ppm. hours, suggesting a higher sensitivity for mature trees.

5. For changes in the composition of species mixtures, the experiments are of limited duration, and there is uncertainty about long-term effects of ozone.

6. In certain studies plants were exposed to other pollutants in addition to ozone. These studies have produced confounding results. More work is needed in this area.

In spite of these uncertainties, the experts preferred to recommend WHO Guidelines on the basis of Critical Levels for which a sound scientific basis existed, rather than incorporate uncertainty factors at that point in time. At least two studies are currently looking at how to incorporate some of the factors (the so-called Level II Critical Levels approach), but these were not far enough advanced to be considered for incorporation into the WHO Guidelines. So the simple Level I Critical Levels fulfil the primary aim of air quality guidelines in providing the best available scientific basis for the protection of vegetation from significant effects. Hence, Critical Levels can provide a solid information basis for risk assessments. Current measurements in rural areas of Europe document that ambient ozone levels substantially exceed the critical levels for crops and trees over large areas. This shows that a high risk of ozone damage to vegetation may exist in Europe.

Table 3.3: The following Guideline Values for Ozone have been Recommended by WHO (WHO, 1999b)

Vegetation type	**Guideline (AOT40) (in ppm.h)***	**Time period****	**Constraints**
Crops (yield)	3	3 months	-
Forests	10	6 months	-
Semi-natural vegetation	3	3 months	-
Crops (visible injury)	0.2 0.5	5 days 5 days	Humid*** air conditions Dry*** air conditions

* AOT40 = Accumulated exposure over a threshold of 40 ppb. The AOT40 is calculated as the sum of the differences between the hourly ozone concentrations in ppb and 40 ppb for each hour when the concentration exceeds 40 ppb, using daylight hours only, i.e. only during hours when cloud-free global radiation is higher than 50 W/m2.

** The appropriate period is selected according to the growing season of the selected sensitive receptor and the climate in the specific region; if it exceeds 3 or 6 months, the most sensitive periods should be used.

*** Related to mean daytime (09.30-16.30) vapour pressure deficit: Below 1.5 kPa means "Humid", otherwise "Dry".

3.3. Exceedance of the WHO guidelines

The data reported under the current ozone Directive are not expressed in terms of the new WHO guidelines. Consequently, guideline exceedance cannot be derived from those data. In what follows, measurement results from the EMEP network (Hjellbrekke, 1997) and concentrations expressed in terms of AOT calculated by the EMEP model (Simpson *et al.*, 1997) are used to provide a preliminary assessment. N.B. Results from the EMEP model are also used in the work of Amann *et al*. (1999) to support the development of the Ozone Reduction Strategy Development and the UN-ECE emission reduction protocols.

The discussion continues as to whether or not an AOT-type statistic is suitable for assessing exposure of the population to exceedance of the WHO guideline value of 120 $\mu g/m^3$ (60 ppb). AOT60=0 ppb. h is mathematically equivalent to zero exceedance of the WHO guideline. The WHO/UN-ECE workshop on 'health effects of ozone and nitrogen oxides in an integrated

assessment of air pollution' agreed that AOT60 could be used as an exploratory indicator for exceedance of the WHO threshold.

3.3.1. Number of exceedances of 120 µg/m3 and AOT60 - human health

Figure 3.1 shows the number of days that the WHO guideline of 120 µg/m^3, calculated as an 8-h floating mean, was exceeded. The data used cover the April to September 1995 period. Note that the EMEP network is composed of rural and remote stations, which makes it somewhat less suitable for assessing human exposure. The figure shows that exceedances occur in all Member States, ranging from 1-5 exceedance days in Sweden and Finland to more than 40 in the north of Italy, Spain and Austria. A calculation of the AOT60 accumulated parameter (Figure 3.2) from the same measurement data reveals a somewhat different pattern. In this case there is a maximum exceeding 5 ppm. h stretching from the south of the UK, across northern France and Belgium to Germany.

AOT60 was also computed by the EMEP model (not shown). The modelled results were combined with maps from geographical information systems. This calculation indicates that 99% of the population in the Community may be exposed, at least once a year, to exceedance of the WHO guideline. Close to 80% of EU15 inhabitants are modelled to experience an AOT60 of 1 ppm. h, with nobody exposed in Finland and 100% exposed in Germany, France and the Benelux countries.

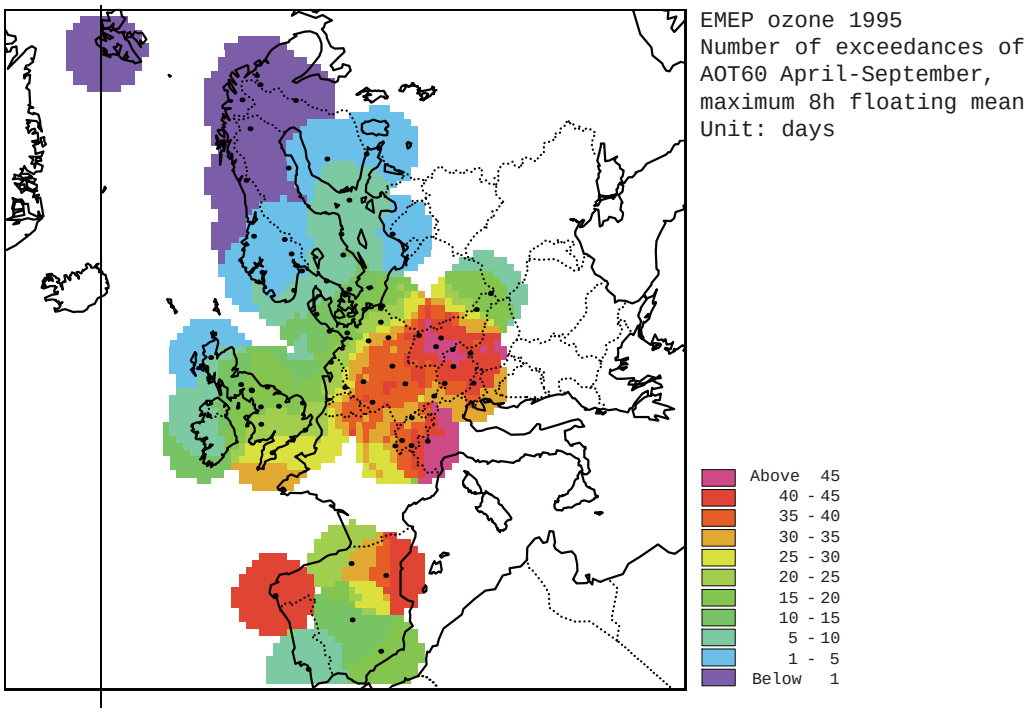

Figure 3.1. The number of days (April-September, 1995) that the maximum 8-h floating mean exceeded 120 µg/m^3. Courtesy: A.-G. Hjellbrekke and the EMEP programme.

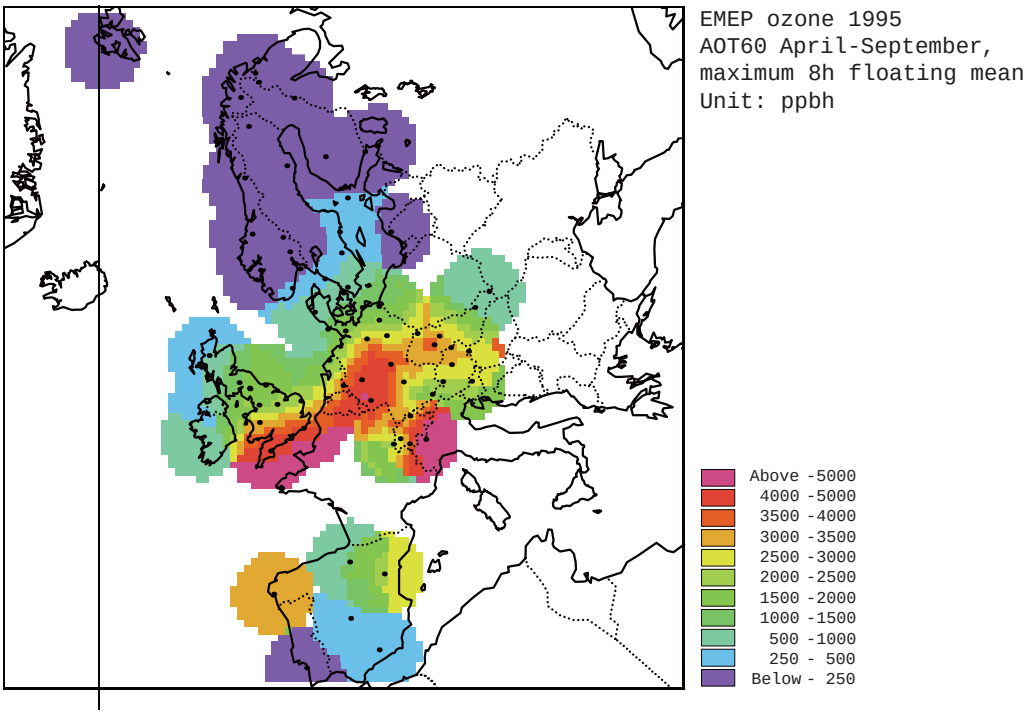

Figure 3.2. AOT60 (April-September, 1995). Courtesy: A.-G. Hjellbrekke and the EMEP programme.

3.3.2. AOT40 - Crops and Ssmi-natural vegetation

The measured and modelled exceedances of AOT40 crops are shown in Figure and 3.4. The modelled map (Simpson *et al*., 1997) is used to enlarge the spatial coverage over all EU Member States. The computed AOT40 levels for crops and semi-natural vegetation are presented as a mean over 5 years so as to remove most of the inter-annual fluctuation induced by meteorology. The calculations are performed using the latest version of the EMEP 150 km Lagrangian ozone model. Emissions were held constant at 1990 levels, but meteorological data were taken from 5 summers, i.e. April-September 1989, ‘90, ‘92, ‘93 and ‘94. This procedure prohibits a one-to-one comparison of the measured and modelled patterns, although the large scale gradient and the location of maxima and minima display reasonable agreement. Both charts show that the threshold of 3 ppm. h is exceeded in most Member States. However, parts of Sweden, Finland and the north of the UK are free from exceedance. GIS calculations based on the computed map indicate that in these countries 44%, 100% and 15%, respectively, of their arable land is protected from exceedance. The calculations also reveal that as an average over all EU15 countries only 6% of arable land is not exposed to exceedance of the AOT40 guideline. In particular French, German, Belgian and Italian crops are calculated to experience exceedance of a factor of about 5 above the 3 ppm. h threshold. Similar exposure patterns for semi-natural vegetation are not available.

The EMEP model indicates that the inter-annual variation of single-year AOT40 calculations is quite considerable; 1985 and ‘86 were ‘low’ ozone years, whereas during ‘89 and ‘90 one episode after the other occurred. The model computed that no matter which year, Sweden, Finland and Scotland have AOT40 levels below 3 ppm. h. In the other Member States, even in the low ozone years, the area of exceedance covers almost their entire surface. The main difference is the magnitude of exceedance: a factor of 2 to 3 in low ozone years and a factor of 5 to 8 in episodic years.

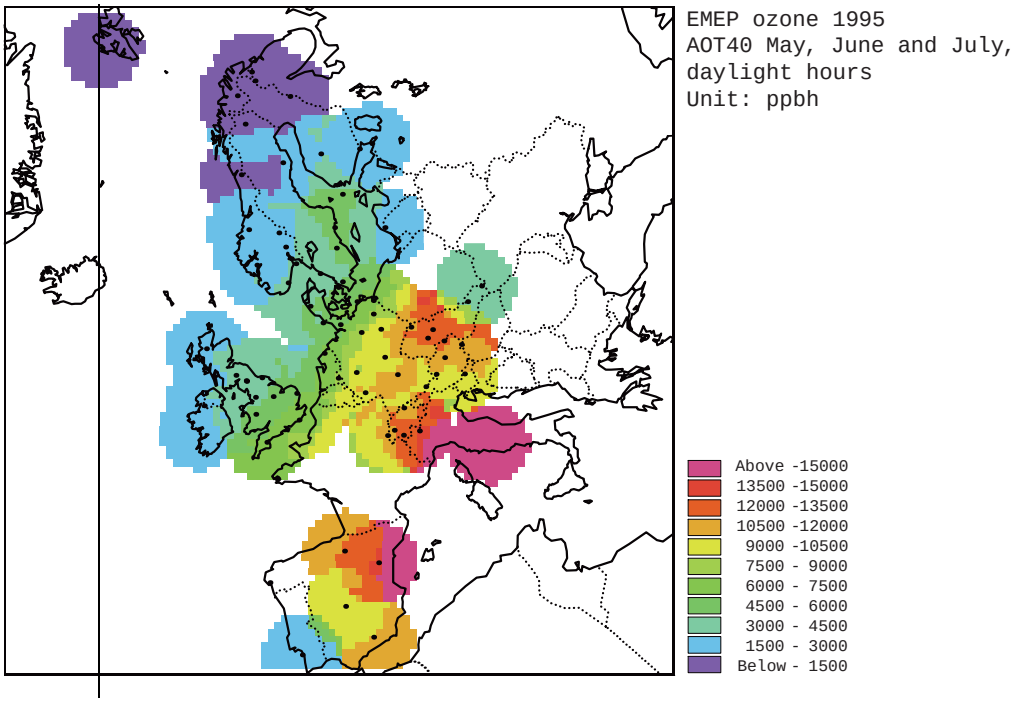

Figure 3.3. Measured AOT40-crops and semi-natural vegetation (1995) calculated from the EMEP network. The WHO guideline is set at 3 ppm. h. 1 ppb O_3 = 2 µg/m^3. Source: Hjellbrekke, 1997.

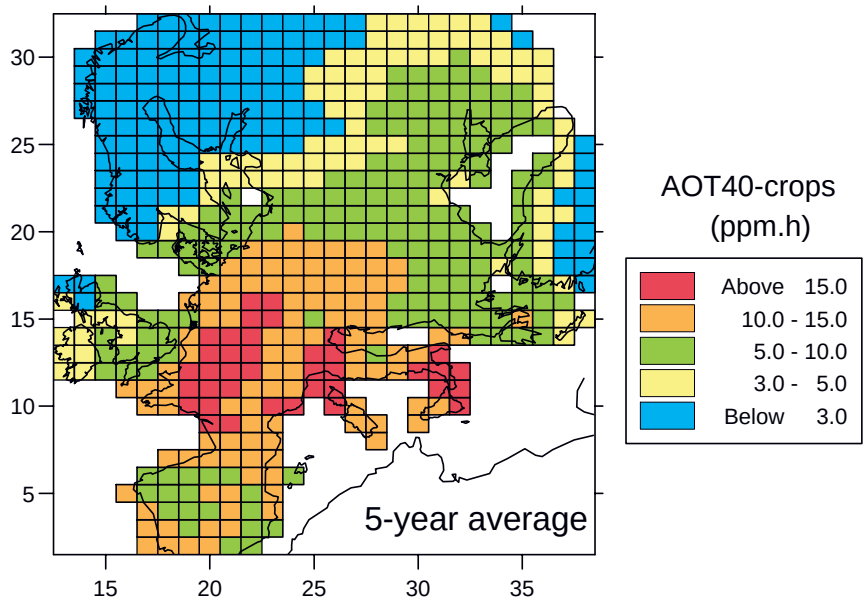

Figure 3.4. Modelled AOT40-crops and semi-natural vegetation as a mean over 5 summers of meteorology using 1990 emissions. The WHO guideline is set at 3 ppm. h. 1 ppb O_3 = 2 µg/m^3. Source: Simpson *et al.*, 1997.

3.3.3. AOT40 – Forests

The 5-year averaged modelled excess ozone over the guideline for forests (10 ppm. h) is presented in Figure. The map derived from the EMEP measurement network is not shown. The modelled figure reveals a Northwest to Southeast increasing gradient. In Scandinavia, Ireland and the United Kingdom forests are almost fully free from exceedance. The Northwest part of the continent

experiences exceedance of the 10 ppm. h guideline by a factor of 2, while some areas of Central Europe and the south of Europe are exposed to a maximum approaching 30 ppm.h. Computations based on data from geographical information systems (GIS) indicate that 35% of Europe's coniferous forest experiences exceedance of the forest guideline. This is in contrast to the broad-leaved forest, of which according to a GIS result approximately 70% is in areas of exceedance. These numbers reflect the large amount of coniferous forest in the Scandinavian countries and the major share of European broad-leaved trees in Germany, France and Italy.

The AOT40 guideline for forests is found to be less stringent than AOT40 for crops and semi-natural vegetation: the area of exceedance is smaller.

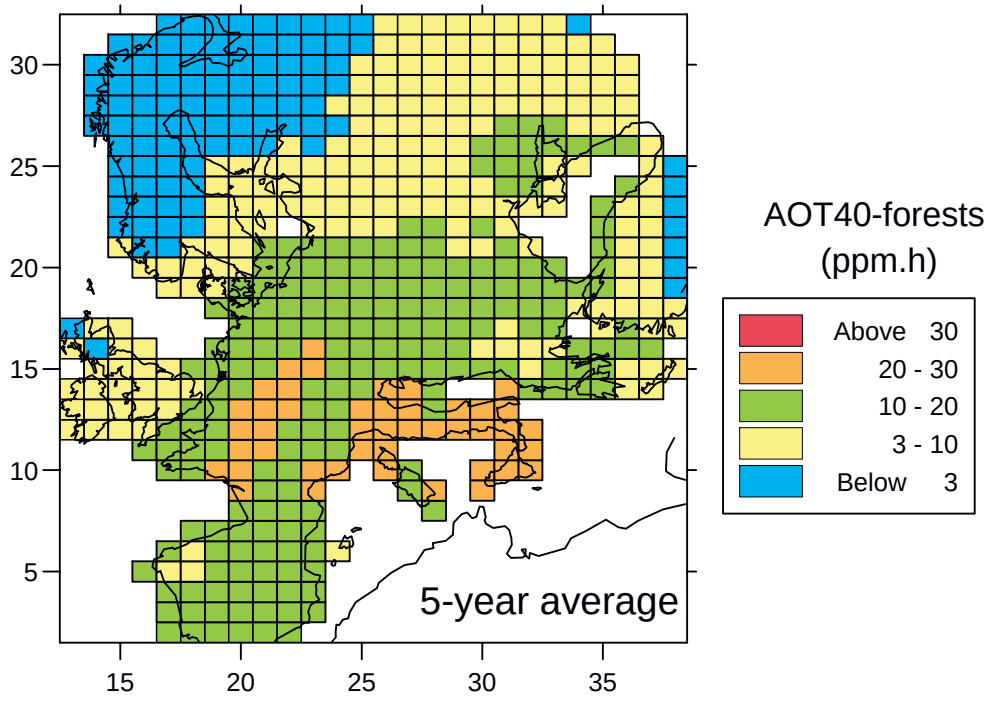

Figure 3.5. Modelled AOT40-forest as a mean over 5 summers. The WHO guideline is set at 10 ppm. h. 1 ppb O_3 = 2 µg/m^3. Source: Simpson *et al.*, 1997

3.4. Existing EU thresholds

Directive 92/72/EEC on Air Pollution by Ozone sets the following air quality thresholds:

1. Health protection threshold

- 110 µg/m^3 for the mean value over eight hours,

which should not be exceeded if human health is to be safeguarded in the event of prolonged pollution episodes.

2. Vegetation protection thresholds

- 200 µg/m^3 for the mean value over one hour
- 65 µg/m^3 for the mean value over 24 hours,

beyond which vegetation may be affected.

3. Population information threshold

- 180 µg/m^3 for the mean value over one hour,

beyond which there are limited, temporary effects on human health in the event of short exposure of particularly sensitive sections of the population and at which the public must be informed by the Member States, including health advice to the population concerned .

4. Population warning threshold

- 360 µg/m^3 for the mean value over one hour,

beyond which there is a risk to human health in the event of short exposure and at which the people must be warned by the Member States, as laid down in the existing Directive.

3.5. Existing standards in Member States

Table 3.4 below gives an overview of ozone thresholds used in the Member States in addition to the protection and information thresholds in the current ozone Directive 92/72/EEC.

Austria

A Federal Act on Measures to Avert Ozone Pollution and to Inform the Public of High Ozone Pollution Levels came into force in 1992. The Act contains ozone warning limits for the purpose of protecting human health against short-term high ozone pollution levels. These values are defined as running three-hour averages. The actual levels are given in the table above.

If the limit value of the early warning level is exceeded at a minimum of 2 measuring stations within an ozone monitoring area, a warning will be issued. The warning includes instant public information and certain recommendations. Warnings must be issued for an ozone monitoring area if it is expected that the warning limit value might be exceeded during the following 24 hours. This must be the case if a value of 260 µg/m^3 as 3-hour mean value (for Warning level I) or 360 µg/m^3 as 3-hour mean (for Warning level II) has been exceeded at, at least two stations in an ozone monitoring area and the concentration level might persist or rise during the following 24 hours. The issue of warnings before the actual limit value (300 and 400 µg/m^3, respectively) is exceeded was regarded as necessary, since it was believed that active measures foreseen in the Ozone Act need time to be enforced effectively.

Additionally, the Federal Act on Ozone prescribes continuous public information on ozone levels during the summer half-year period, irrespective of currently measured ozone values.

The Air Quality Protection Act, which entered into force on 1 April 1998, sets a target value of 110 µg/m^3 as an 8h mean, defined according to the Ozone Directive 92/72/EEC.

France

Since April 1997 a decree has been in force in Paris which specifies that if ozone 1-hour mean concentrations exceed or are anticipated to exceed 360 µg/m^3 the following day at 2 or more sites, a partial traffic ban should be established in the French capital and its immediately neighbouring cities. During this period cars having either odd or even licence numbers are not allowed to run; several categories are excluded from this ban. Public transportation is then free in the relevant areas.

Germany

On 19 July 1995 an amendment of Article 40a of the Federal Immission Control Act ("Ozone Law") was adopted, banning highly polluting motor vehicles (except heavy duty vehicles) from the

roads on the day following the first occurrence of high ozone values. It has to be announced in the affected Federal States if a 1h-mean concentration of 240 $\mu g/m^3$ or more is recorded at a minimum of three monitoring stations, to be more than 50 km and less than 250 km apart from each other, and if it can be assumed that this will occur again on the following day. When the EU information threshold of 180 $\mu g/m^3$ is reached, the authorities must request people not use their cars.

Italy

In 1983 a Decree was approved to set National Ambient Air Quality Standards (NAAQS) with a view to ensuring the health of the population and "public welfare". The Decree established that implementation of these standards was the responsibility of the regions. The NAAQS for ozone was 200 $\mu g/m^3$ as a 1-hour maximum concentration not to be exceeded more than once per month.

A step forward in the legislation process was made in 1992 when a Ministry of the Environment Decree on urban pollution was enacted. An information level and an alarm level for ozone of 120 $\mu g/m^3$ and 240 $\mu g/m^3$, respectively, were adopted.

In 1994 a subsequent Decree established an information level and an alarm level of 180 $\mu g/m^3$ and 360 $\mu g/m^3$, respectively.

In 1996, to comply with the requirements of Directive 92/72/EEC, Italy enacted a Decree of the President of the Republic (16/05/96) which integrated the Directive and the corresponding NAAQS into Italian legislation. Article 5 of the Decree requires the implementation of measures to counteract a status of alarm (i.e. beyond the warning threshold) when the information level has been exceeded for three consecutive days and when meteorological conditions are not conducive to the improvement of the situation.

Sweden

Swedish legislation has no limit or standard values for ambient ozone concentration additional to the information and warning threshold values of the EU Directive. However, the Swedish Environmental Agency has national targets to protect environment and health. For the protection of human health the ozone concentration of 120 $\mu g/m^3$ (1-hour mean) should not be exceeded more than 12 times per year, and 150 $\mu g/m^3$ (1-hour mean) should not be exceeded at all. For the protection of ecosystems, the mean value over the growing season (April-September, 9-16h CET) should not exceed 50 $\mu g/m^3$.

Recently the Swedish EPA proposed 80 $\mu g/m^3$ as the maximum 1-hour mean level as a new guideline to protect human health. It also proposed to adopt the new UN-ECE critical levels for crops and for forest trees (see Section 3.2.2)

United Kingdom

In March 1997 the UK National Air Quality Strategy was published. This contained a series of health-related standards, as benchmarks for air quality, based on the work of the Expert Panel on Air Quality Standards. The Strategy also contained air quality objectives, which policies aim to achieve. The provisional objective for ozone is 50 ppb (100 $\mu g/m^3$) as a daily maximum running 8-hour mean, to be exceeded no more than ten times per year. The Strategy recognised however that this objective could not be achieved by measures in the UK alone and that international action would be required.

Table 3.4: Ozone thresholds in Member States in addition to existing EU thresholds.

Member State	Ozone thresholds
Austria	Federal Act on Ozone: Early warning level 200 µg/m^3 as running 3 hourly means Warning level I 300 µg/m^3 Warning level II 400 µg/m^3 if exceedance occurs at 2 stations or more Federal Air Quality Protection Act: Target Value 110 µg/m^3 as 8h average (same definition as in Dir.92/72/EEC)
Belgium	None
Denmark	None
Finland	None
France	None
Germany	Maximum Pollution Concentration ("Immission") Values: Protection of human health: 120 µg/m^3 (0.5h mean), below which adverse effects were certainly excluded. For protection of vegetation: Concentrations in µg/m^3 *Exposure duration* *Resistance level* 0.5h 1h 2h 4h 8h Sensitive 320 160 110 90 70 Intermediate: 480 320 240 190 160 Less sensitive: 800 480 400 370 320
Greece	None
Ireland	None
Italy	None
Luxembourg	None
The Netherlands	Non-legal limit values for the year 2000: • 240 µg/m^3 (1 hour), permitted exceedance max. 2 days per year • 160 µg/m^3 (8 hour), permitted exceedance max. 5 days per year • 100 µg/m^3 (growing season May - September, 10.00 – 17.00 hour) Guide value for the longer term: no exceedance of the above 1h and 8h levels Ultimate goals: • 120 µg/m^3 (1-hour) no exceedance • 50 µg/m^3 (growing season average)
Portugal	None
Spain	None
Sweden	National Target Value: For protection of human health: the concentration of 120 µg/m^3 (1h mean) of ozone should not be exceeded more often than 12 hours per year and the level of 150 µg/m^3 should not be exceeded at all. For protection of the environment: the mean value over the growth season (April-September 9 a.m. - 4 p.m.) should not exceed 50 µg/m^3.
United Kingdom	Ozone standard: 50 ppb as 8h daily maximum. Provisional Objective to be achieved by 2005: 97th percentile level of the standard, i.e. not to be exceeded on more than 10 days per year

3.6. Standards in countries outside the European Union

Japan

The hourly environmental standard is set at 60 ppb for photochemical oxidants (120 µg/m^3 for ozone). When concentrations of photochemical oxidants reach an hourly value of 120 ppb (240 µg/m^3) and it is apparent that meteorological conditions favour a continuation of this condition, an alert is issued and a variety of measures are taken to prevent damage to health, such as discouraging outdoor exercise.

Switzerland

The Clean Air Ordinance contains Pollution ("Immission") Limit Values which would, if attained, provide protection from adverse effects in general. The ozone limit values are 120 µg/m^3 (1h-mean) to be exceeded not more then once a year and 100 µg/m^3 as 98-percentile of the 0.5h values within a month

USA

Currently the primary (health related) and secondary (vegetation related) USA national ambient air quality standards are both set at 0.12 ppm (240 µg/m^3) as 1-hour average. An area attains (is in compliance) when the number of days per year on which the level is exceeded is less than or equal to 1, averaged over 3 years.

After extensive reviews of new scientific findings, EPA concluded that the current standard is not sufficient to protect the public and vegetation from adverse effects (US-EPA, 1996b). Recently (25.6.1997) President Clinton approved the EPA proposal to replace the current primary and secondary standard by an 8-hour standard set at 0.08 ppm (160 µg/m^3); an area would not attain when the 4th highest daily maximum 8-hour concentration, averaged over 3 years, is above 0.08 ppm.

3.7. Values to be taken as starting points for setting EU Air Quality thresholds

In Chapter 2 a two-level approach is proposed, leading to the definition of a long-term objective and a target value. Before numerical values for any of these air quality thresholds for ozone can be proposed, the definition of the threshold parameter should be chosen first: maximum, percentile, and/or AOT. The parameters as recommended by WHO will be taken here as starting point for the discussion: the 8 hour average concentration for health protection, and AOT40 for ecosystem protection.

Proceeding from this starting point, pragmatic adjustments in the definitions have to be made in order to make it suitable for assessment in a monitoring network. One should note that there is an important difference between the application of these definitions in the framework of the Daughter Directive, which aims at air quality targets, and for ozone strategy calculation such as carried out in the framework of the UN-ECE negotiations on a new multi-pollutant and multi-effect protocol and the drafting of a NEC Directive, which aim at emission targets. For UN-ECE, an exact definition of the air quality targets is less essential than for the Daughter Directive. From the perspective of the Daughter Directive, an unambiguous definition of the air quality targets is essential for monitoring the air quality and assessing compliance. Current Modelling limitations which mean that only certain air quality target definitions/units can be modelled directly, should not be regarded as a major reason to reject an alternative definition/unit; the model is in almost all respects an approximation of reality.

Format of the target value for human health

An important matter to consider is the question whether environmental objectives should be defined as a maximum value (no exceedances allowed) or as a percentile, i.e. a value that is exceeded by a specified number of times per year (or in several years). If taking a percentile, then there is the question of choosing a higher percentile with lower exceedance or vice versa.

From the health effect point of view as such, there is no reason to allow exceedances. For the general public, a threshold expressed as a level that is allowed to be exceeded several times is more difficult to understand than a maximum allowed value. This is certainly true for setting the Long-term Objective, as this is directly based on the WHO guideline.

Concerning the target value, it was proposed above in chapter 2 to set a target value as an interim objective to be attained as far as possible by the year specified. This requires fixing the target value somehow, at an intermediate level above WHO. Two possibilities have been suggested:

1. a target value based directly on the WHO guideline for the protection of human health i.e. to express it as 120 μg/m³ not to be exceeded more than a certain number of days per year, say 20. This has the advantage of transparency with respect to the WHO guideline. The target value is designed to reduce exceedances of the guideline from today's high level to a much lower level as soon as possible. Public information will be linked in part to the target value and this formulation would show most clearly how far strategies are succeeding. A potential disadvantage is that the number of allowed exceedances is relatively high. Attention would be focused on continuing exceedances of the WHO guideline in areas where ozone levels are relatively high, which could lead to a perception of failure even once exceedances are substantially reduced, and give rise to public concern, even though the majority of the public should not notice significant effects during exceedances. EU environment ministers however agreed a similar construction for this in the Common Position on the proposal for a Directive setting limit values for SO_2, NO_x, PM_{10} and lead and there is a precedent in national standards for ozone in the UK;

2. a target value expressed as a higher concentration, with a smaller number of allowed exceedances, for example 3 or 4. 160 $\mu g/m^3$ has been suggested during discussions of the Working Group as a concentration at which clinically significant effects appear in active, healthy subjects (young adults and children) in controlled tests. (It is also the concentration in the new US Ambient Air Quality Standard for ozone.) Such a formulation would be closer to the format of most ambient air quality limit values. It might therefore be more easily understood and might command more public confidence. This formulation obscures however the link to the WHO guideline of 120 $\mu g/m^3$. There is a danger that the proposed target value would appear arbitrary, or would appear to be an attempt to hide information with respect to the WHO guideline. If this were so it would command less public confidence than the formulation showing clearly its basis in the WHO guideline.

In making a choice the primary consideration should be whether one formulation might better reduce risk to public health than the other. For example, it might be argued that a target value of 160 $\mu g/m^3$ with few exceedances would be more effective in reducing peak ozone concentrations than a target value of 120 $\mu g/m^3$ with a higher number of exceedances. If high peak concentrations were the main concern it could provide better protection. There are two aspects to consider: whether in air quality terms it is likely that a target value of 120 $\mu g/m^3$ would be likely to result in higher peaks that one of 160 and whether if so, this would be significant in health terms and in terms of the number of individuals likely to be affected by those significant health effects.

Where air quality is concerned, the proposed measures and emissions reductions in the ozone strategy and proposed national emission ceilings directive (NEC) will ensure that the target value is met as far as possible at regional level. The target value is indeed derived from consideration of the measures available to comply with the NEC proposal (see chapter 5.1). The NEC Directive,

and therefore regional ozone levels, will not change however the target value is expressed. However, further local action may be needed to reduce locally generated exceedances. The formulation of the target value could therefore be important at local level. But it has been shown that measures available to reduce ozone levels towards 120 μg/m³ will reduce peak ozone concentrations more rapidly than lower concentrations[14]. Consequently, the first formulation should not in practice allow a large number of high peaks in ozone concentrations.

It is nevertheless useful to consider whether it is more important in health terms to concentrate on reducing high peaks of ozone or to concentrate on reducing ozone levels more generally. As stated above in section 3.2, whilst controlled studies do find thresholds for the appearance of effects in laboratory studies using healthy adults as subjects, there is evidence for a linear relationship between exposure in the environment and health effects on the general population. The relationship may hold at concentrations down to and below 120 μg/m³. That is, an increase in concentration from 120 to 140 μg/m³ would have the same increment in effects on health as an increase from 160 to 180 μg/m^3. Therefore the view of the overwhelming majority of the Working Group is that its is at least as important to reduce frequent variation at lower concentrations by reducing average concentrations, as it is to reduce relatively infrequent high peaks. Indeed it may well be more important.

In fact, in developing their proposal for the recently introduced U.S. Ambient Air Quality Standard of 160 μg/m^3 as an 8 hour, the EPA observed[15] that this standard "*does not necessarily reflect a threshold below which effects do not occur, but rather reflect(s) levels at which studies finding statistically significant effects of concern have been conducted.*" The EPA also assessed risk at concentrations below 160 μg/m^3 and concluded that a lower level of 140 μg/m^3 "*would provide increased protection from long-term exposures that may be associated with potentially more serious but more uncertain chronic effects*". Further evidence as to the effects of ozone at lower concentrations than 160 μg/m^3 has accumulated since the EPA reached this conclusion.

The Working Group on ozone has considered the advantages and disadvantages outlined above. A large majority of the Group is of the opinion, on grounds of risk and transparency, that **the target value should be expressed as 120 μg/m^3, with some (20-25, see section 5.1) allowed exceedances per year.**

There are also strong practical arguments against taking the concentration maximum or a very high percentile (equivalent to only a few exceedances) for environmental objectives that have legal implications (e.g. to implement measures reducing emissions). Of all statistical parameters, the maximum concentration is the most variable one. This variability causes zones where levels are close to the limit value to fluctuate in and out of compliance from year to year. For ozone, meteorological (random) variations cause the annual 8 hour maximum to vary by 10 to 20%, which is large compared to the changes that can be brought about by air quality management in the course of a year. Only if high concentrations were accurately predictable and short-term reduction measures made it possible to 'shave' the peaks, would air quality management be able to cope with such fluctuations. Since fluctuations of the compliance status can not be dealt with satisfactorily by air quality management, one should from the administrative and management point of view, attempt to minimise such fluctuations. Allowing a specified number of exceedances would diminish these problems, because the variability of the compliance rate decreases with the number of exceedances.

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[14] In identifying a cost-effective solution for the NEC proposal the ozone objective used in the optimization (RAINS model by IIASA) was expressed as an AOT60 (see below under "AOT for health") which gives higher concentrations a higher weight.

[15] US-EPA, 1996b, p. 166

A second advantage of lower percentiles (with a higher number of exceedances) is that they are less sensitive to measuring errors or modelling uncertainties than the maximum or higher percentiles with only a few exceedances.

Recently the US-EPA proposed to deal with this in a slightly different way by defining compliance on the basis of three years. The average of the three fourth highest annual 8-hour values is compared with the concentration level specified in the air quality standard. This can be repeated every year using a moving 3-year average providing an annual parameter with a considerably reduced variability. More generally, the fluctuation problem could be reduced by taking a longer time period than one year to determine the implications of exceeding an environmental objective. This could be realised in several ways: the environmental objective itself could be defined over a time span of a specified number of years, or the legal implications could be triggered by a specified exceedance rate over a specified number of years.

To minimise the effect of meteorology on compliance checking and to better visualise the trend of ozone pollution resulting from emission reductions, a majority of the group were in favour of considering the allowed exceedances specified by the target value over a three-year averaging period. With 2010 as the target year for achieving the national emission ceiling there are in principle several possibilities for setting the 3-year averaging period relative to the target year: 2010 could be the first, second or last year of the period. Choosing 2010-12 would result in unnecessary delays in checking compliance. The case of 2008-2010 would not account for the fact that, in deriving the target value, implementation of emission reductions is only assumed for 2010. Therefore it is proposed to centre the 3-year period around the target year for the emission reduction, i.e. to take 2009-2011.

A minority of the Working Group were not in favour of basing the compliance check on a multi-year period, as this would not be transparent with respect to the long-term objective. Moreover, it might be difficult to understand for the public, given that health effects appear largely due to short-term exposure in each year. Applying a multi-year period could delay the implementation of abatement measures triggered by any non-compliance with the target value. The same opinion was expressed regarding the five-year period for the vegetation-related target value.

AOT for health

An option to be considered for setting a health threshold is AOT60, the cumulative exposure above 60 ppb (120 $\mu g/m^3$), on the basis of 8-hour averages. A threshold expressed as a percentile, which allows a specified number of exceedances, counts each exceedance as one, irrespective of the exceedance rate. AOT on the other hand weighs each exceedance by its exceedance rate, which can be expected to correlate better with the risk. This has been discussed before, especially in relation to the strategy development for the second NO_x Protocol in the framework of the UN-ECE Convention on Long Range Transboundary Air Pollution (UN-ECE/WHO, 1997). Other arguments in favour of AOT are similar to those of allowed exceedances: it is more stable than the maximum, and less sensitive to errors and uncertainties.

Compared with percentiles (allowed exceedances), AOT is however more difficult to understand for the public than the number of allowed exceedances. An advantage of AOT compared with the number of exceedances is that it can be directly used in the Integrated Assessment Model that will be used to develop ozone reduction strategies.

Definition of the 8-Hour time window and counting procedure

If no exceedance is allowed, the obvious choice for the 8-hour time window would be to check the entire time series of hourly concentration for the occurrence of any period of 8 hours in which the average concentration is above the limit value, irrespective of the starting time of that period. But if several exceedances are allowed, the question of how the 8-hour periods are to be counted

becomes important. If all running averages were counted, many exceedances would overlap, e.g. there could be 24 exceedances in one day. If overlapping time windows had to be avoided, fixed time windows could be chosen, e.g. 0-8, 8-16 and 16-24h. In many cases, however, this would overlook the highest 8-hour mean of the day, which is typically centred around the late afternoon; this peak period would be divided between two periods. If the time window was not fixed, the procedure for finding the maximum number of exceedances of non-overlapping periods would not be very transparent. The proposal is to choose the method of the daily 8-hour maximum: select for each day the highest daily 8-hour average from the 24 moving averages and test this value against the threshold. This procedure counts the "exceedance days". For a complete specification the assignment of the 8-hour period to calendar days should also be defined: it is proposed to assign each 8-hour period to the day of its last hour (so the period 17-1h is the first 8-hour period of a day) (Directive 92/72/EEC) [16]

Proposal for the health thresholds

Long-term Objective for Health

For reasons of transparency the following long-term objective is proposed for health for protection against acute health effects (e.g. decrements in lung function, asthma exacerbations, hospital admissions):

Long-Term Objective for health: the maximum 8-hour average concentration is set at 120 µg/m^3, no exceedances are allowed. The maximum is calculated from running 8-hour averages of 1-hour mean concentrations.

While it is recognised that chronic exposure to ozone may also produce effects, quantitative data are insufficient to estimate the degree of protection offered by the long-term objective against possible chronic effects.

As a result of the large inter-annual fluctuation of the maximum, major compliance fluctuations are to be expected when the actual levels are close to this objective. The administrative complications that such fluctuations may generate have been a reason to allow other pollutants a certain number of exceedances of the limit value. This is not very relevant for the long-term objective for ozone, since it is not intended to attach important administrative or legal implications to fluctuations in compliance. The implications of inter-annual fluctuations for the assessment requirements can be relaxed by linking these requirements to multi-year exceedance (see Chapter 4).

Target Value for health

For reasons of transparency it is proposed to use the same air quality parameter for long-term objective and Target Value, but to allow a specified number of exceedances for the Target Value. In future, the number of exceedances could be gradually reduced, with zero exceedance (the long-term objective) as the ultimate goal. The compatibility problem with the Integrated Assessment Model is not considered a decisive reason to opt for the AOT parameter form. Specification of the stringency of the Target Value should depend not only on risks, but also on e.g. economic

[16] In the current Directive 92/72/EEC, exceedance of the health related threshold is counted for four fixed 8-hour periods per day (0-9h, 8-17h, 16-1h, 12-21h). The proposed procedure to take each of the 24 8-hour periods into account is somewhat more stringent. A limited analysis of data in the AIRBASE database indicated that a LTO at 120 µg/m3 (considering 24 periods) would allow about 1 or 2 exceedances per year of the current health related threshold at 110 µg/m3 (considering 4 periods).

considerations. This will be addressed later. The following recommendation on the Interim Objective is given:

Target Value for health: the daily maximum 8-hour average concentration is set at 120 µg/m^3; this value may be exceeded on a specified number of days in a certain period. A proposed range of the number of exceedances (20-25 days per year) is given in Chapter 5.1. For checking compliance a three-year period is recommended. The daily maximum is calculated from running 8-hour averages of 1-hour mean concentrations.

3.7.1. Long-term objective and target value for the protection of vegetation

The WHO recommends five thresholds for AOT40 (see Table 3.3): one for forests, one for semi-natural vegetation and three for crops. For crops, the 3-month value refers to yield protection, and the other two to visible injury, based on five days. For visible injury, humid and dry conditions are distinguished.

Given the basis of the definitions of the LRTAP Convention Critical Levels and WHO Guideline values it is not easy to see how these might meaningfully be translated directly into useful long-term objective or target values. Exceedance of the guideline value does not necessarily result in adverse effects, while attainment of such a value does not prevent damage. There is the question whether the degree of damage used to establish the critical level, effectively the minimum statistically observable, is appropriate for a limit or target. Further, the added complexity of averaging over 5 years (to allow for the wide variation in ozone concentrations resulting from weather conditions from year to year) and the definitions of growing season and daylight hours make the applicability of an objective or target difficult to specify for any single region.

Consideration should perhaps also be given to the basis from which the WHO guideline values and Critical Levels were derived. While in the early stages of the development of the Critical Loads and Levels concept it was envisaged that exceedance maps would demonstrate the need for pollution abatement, more recently such maps and threshold values have been used more directly, as input data for integrated assessment models for developing strategies for LRTAP Convention protocols. The simple (Level I) Critical Level values may prove effective for this purpose, as they are being compared with simple atmospheric modelled ozone concentrations and the integrated assessment models use simple abatement cost measures to estimate an overall strategy across Europe. At a local level the realities of exceedance and vegetation damage are very different and are influenced strongly by the complicating factors identified in previous sections. The experts at the UN-ECE workshop in Kuopio in 1996 (Kärenlampi and Skärby, 1996) stated specifically that "*the exceedance of the critical level of 3ppm.hours must not be converted into a yield loss estimate, but it should only be used as an indication of the degree of risk*". Similarly the WHO workshop recommended the guideline values as a "*solid basis for risk assessment*" and the "*best scientific basis for protection of vegetation*".

Although the WHO workshop considered the guideline values a solid basis for risk assessment, there was clear concern about the uncertainties and how the guidelines might be used to derive regulations and abatement strategies. The question remains: what threshold value or values might be considered to provide an adequate level of protection to translate into air quality target values and abatement strategy, respectively? Annex II of the Council Directive on ambient air quality assessment and management provides a list of factors to be taken into account in the setting of Limit Values and alert thresholds, as appropriate. These factors include:

- Climatic conditions;
- Sensitivity of flora and fauna and their habitats,
- Long-range transmission of pollutants; and
- Economic and technical feasibility.

Experts involved in advising the WHO have clearly identified that variations in climate, vegetative species, topography, etc., are significant in determining the risk of ozone damage to vegetation across Europe. In addition, there are important transboundary and long-range transport issues to be considered, most notably the contribution to ozone concentrations of emission sources outside the EU (e.g. Eastern Europe) and beyond national boundaries within the EU; and trends in global emissions and global background concentrations of ozone (e.g. contribution from North America). Naturally occurring variations in ozone concentrations (e.g. with altitude), the relative magnitude of the impact of other plant stresses, the role and cost-effectiveness of alternative risk management methods and the compatibility of any approach with other EU or UN-ECE initiatives will also need to be considered. All these issues will influence the economic and technical feasibility of any approach that is taken to abate ozone and protect vegetation, and need to be considered alongside the guidance provided by WHO.

One could consider the possibility of applying the guideline for each vegetation type only to the air quality above that vegetation type. This would mean that the threshold would have a spatially differentiated pattern, coinciding with land use, which would make assessment (compliance checking) rather complicated. This spatial differentiation could be decreased by choosing large averaging areas. If the guideline for crops and semi-natural vegetation (3 ppm.h in 3 months) is the most stringent guideline, the averaging areas could be made so large that this vegetation was always present, so that this threshold could be chosen as a starting point.

So, assuming conditions to apply to (very) large areas instead of making local differentiations could provide possibilities.

The contribution of global background levels to AOT40

On non-episodic days the global background level is an important contributor to ozone concentrations. Consequently, air quality parameters that are not exclusively sensitive to the high episodic levels of ozone are sensitive to the global background. The average background ozone near the sea surface before air masses move into Europe from the west is 30 to 35 ppb at European latitudes (P. Borrell, et al., 1997), and seasonal and shorter term variations give rise to a large contribution to AOT40. This contribution is large because the number of hours with concentrations slightly above 40 ppb is much larger than the number of hours with levels close to the maximum concentrations. Trends in the global background will therefore be reflected in AOT40 values. Currently, background stations at European latitudes do not show a clear trend, but most scientist believe there was a large upward trend before 1980, due mainly to growth in anthropogenic sources. If, due to increasing emissions in developing countries, the future trend were to be upward, it might offset benefits in the AOT40 value produced by European efforts to reduce episodic ozone levels. On the one hand one may argue that this is not relevant, because the global contribution to exceedance of AOT40 is due to man-made sources; on the other hand the global contribution is difficult to control at the EU level. A higher reference level, e.g. AOT60, would be less sensitive to the global background and could be chosen to be approximately equivalently strict. However, choosing this level would weaken the relationship with the WHO guideline.

Inter-annual variability of AOT40

Occurrence of high ozone levels varies greatly from year to year and consequently the number of exceedances of 40 ppb fluctuates markedly. Since AOT essentially weighs each exceedance by its rate of exceedance, which is also variable, AOT fluctuates even more than the number of exceedances. This variation is largely due to meteorological fluctuations between years and is beyond the control of air quality management. It is not useful, therefore, to base structural reduction measures on each year alone. By judging exceedance over a multi-year period, fluctuations can be decreased.

If a multi-year approach were opted for, assessment could be based on an unfavourable year in the multi-year period (e.g. the worst) or on a mean (or median) for the entire period. If the worst year were taken, the fluctuation problem would not be remedied, and the disadvantages would remain. So, the parameter to be judged should be a mean (median, etc.) one. This would mean that high ozone levels within a single year would be considered acceptable as long as the exceedance rate allowed for the multi-year period is not exceeded. Also, a multi-year period for judging compliance would increase the feedback time between air quality developments and policy, but this is only an illusory drawback, since real-life conditions do not allow significant ozone trends to be detected in one or two years. It is proposed to judge the threshold for vegetation on a five-year average basis.

Proposal for the threshold

The WHO guideline for crops and semi-natural vegetation is in practice virtually always more stringent than the guideline for forests. It does not seem likely that this will change, though the future frequency distribution of ozone may be somewhat different from the current. Consequently, the guideline for crops and semi-natural vegetation is also expected to provide sufficient protection for forests, and the guideline for forests will not be taken separately as a basis for the environmental objectives.

The guideline for visible injury is even more stringent than AOT40 for crops and semi-natural vegetation. Consequently there are grounds for considering it as a potential long-term objective. It does, however, not seem justifiable to replace the AOT40 for crops and semi-natural vegetation, since the purpose of preventing visible injury is primarily to avoid economic damage, while the guideline for crops and semi-natural vegetation has a broader scope. The practical possibilities of measuring exceedance of the guideline for visible injury are very limited because of the dependence on the vapour pressure deficit. Since, in addition, the dependence on vapour pressure deficit is uncertain, it is proposed not to set a target value or a long-term objective for visible injury. It is not expected that such objectives would lead to an ozone reduction strategy that is different from a strategy based on the Target Value for crops and semi-natural vegetation. It is proposed to include 5-day AOT values in the reporting requirements (see Chapter 6).

Long-term Objective for vegetation

Based on the above considerations, the following long-term objective is proposed for vegetation:

Long-Term Objective for vegetation*: *AOT40 (daylight hours; 3 months) is set at 3 ppm.h. The daylight and 3-month time windows are specified below.

Target Value for vegetation

For reasons of transparency it is proposed to use the same air quality parameter for the long-term objective and Target Value, but to set the Target Value a at higher value. In the future, this value could be gradually reduced, with 3 ppm.h (the long-term objective) as the ultimate goal. The following recommendation on the Target Value is given:

Target Value for vegetation: AOT40 (daylight hours; 3 months); a proposed range (8-9 ppm.h) is given in chapter 5.1. For checking compliance a 5-year average is recommended.

3-month window

In the WHO guideline for crops and semi-natural vegetation, which is based on the UN-ECE critical levels, it is specified that a 3-month period should be selected according to the growing season of the selected sensitive receptor and the climate in the specific region; if the growing season exceeds 3 months, the most sensitive period should be used. The WHO definition does not

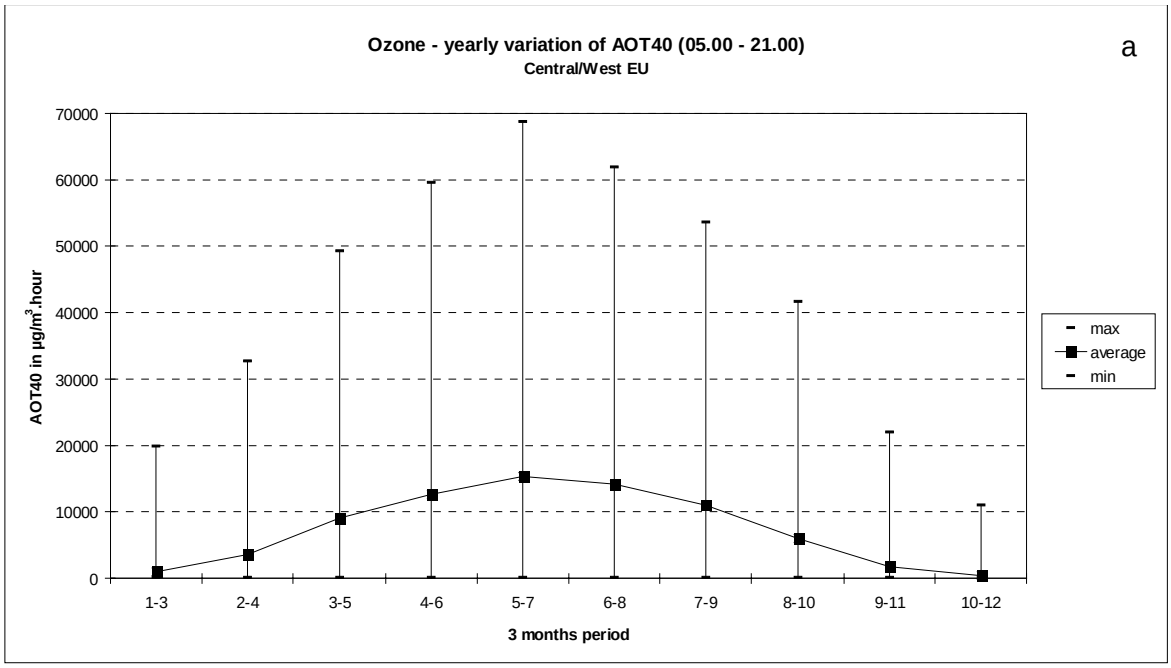
take account of spatial variability in the length of the growing season, but fixes the AOT40 value for crops and semi-natural vegetation at 3 months.

The main spatial variability of the growing season is in the North-South direction, though it must be borne in mind that vegetation species are not distributed uniformly across the EU. To make the WHO definition operational, one has to select the appropriate sensitive receptor species for each EU region, determine its growing season and select the corresponding 3-month period. This difficult work has so far not been done, and so a reference to compare simpler definitions of the 3-month period with is not available.

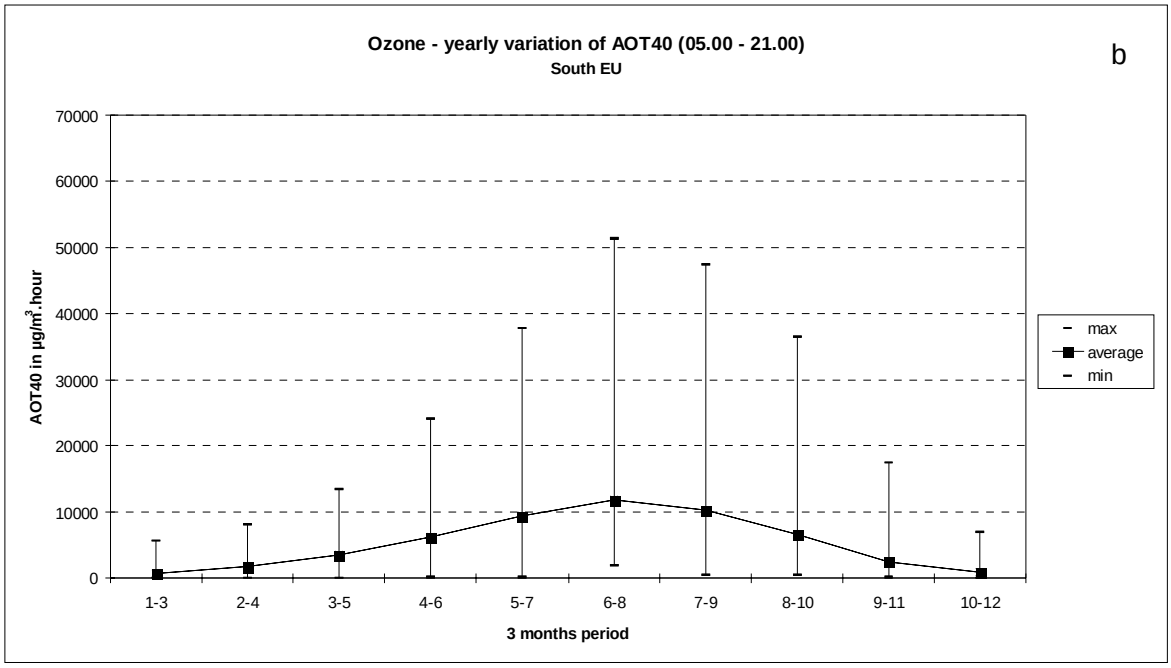
For modelling purposes, where a uniform window was desired, UN-ECE recommended the 3-monthly period of May-July.

An analysis of data from the European database APIS showed that in Central/Western Europe AOT40 for the period May-July was on average somewhat higher than for April-June or June-August (Figure 3.6). In Southern stations the highest average was found for the period July-August (Figure 3.7), but for stations in the Valencian Region this period was found to be May to July[17]. For Northern Europe very few data were available; these indicated that the highest contributions occur in spring. The choice of period should, however, be determined not by the seasonal pattern of AOT40, but by the growing season or the period of highest sensitivity of the vegetation, which is currently undefined.

Similar to the situation for the daily window, a uniform window per Member State or for the EU as a whole could be an option. In view of the many uncertainties **it is proposed to choose the simplest option, in which the growing season window is taken uniformly over the EU, identical to the window proposed for modelling purposes, i.e. May-July.**



[17] Information supplied by J. Sanz, CEAM, Valencia, Spain.



Figures 3.6 and 3.7. Yearly variation of AOT40 (3-month period) for Central/West and South EU, based on APIS data.

Daily window

The WHO guideline for crops and semi-natural vegetation specifies that exceedance of 40 ppb should be counted in the calculation of AOT40 only when the cloud-free global radiation is higher than 50 W/m^2. This defines a daily window that is independent of weather. For practical purposes it can be regarded as depending only on the position on the globe and the day of the year. When related to the time on the clock instead of solar time, the time window is different everywhere. From the East to the West of the EU there is a difference of 42° longitude, equivalent to 42/15 = 2.8 hours in local solar time. Large differences also exist between the North and the South of the EU, while in the extreme North, above the polar circle (67°), the sun remains above the horizon 24 hours a day in summer. In the South, daylight does not last much longer than 12 hours. In the North the duration is longer; since the sun is low in the sky there, the threshold of 50 W/m² is seen to be important: even far above the polar circle the duration of the window is less than 24 hours in mid-summer.

A simple definition is required, both for operational reasons and for transparency. It is important to note that the WHO definition is also a very simplified approach (the so-called Level I), which takes no account of many variations in sensitivity of vegetation across the EU. Two simplification steps are proposed:

1. The time window is defined in terms of whole hours: the time window is rounded to the nearest whole hour (xx.00 hour). This has clear operational advantages.

2. A uniform time window per area is chosen. The larger this area, the larger the deviation from the WHO definition in the border regions of the area. Uniformity could be applied per individual Member State, thus limiting the deviation. This would mean that the time window would change at the border between Member States, which would produce some discontinuities in the AOT40 pattern. A larger step would be to define uniformity per time zone (there are three in the EU). This would limit the deviation from the original WHO guideline in an East-West direction to somewhat over half an hour, but the full North-South deviation would still exist. The largest step would be to apply a uniform time window for the EU as a whole. This would mean a deviation from the WHO definition of about 1.4 hours at the East/West borders, and several hours at the North and/or South

border of the EU, depending on how the window is situated in time. The obvious advantage is the simplicity of the choice. There would be no discontinuities in AOT patterns at the EU's internal borders, though the discontinuities at the outer (Eastern) border would be large. An additional advantage would be that the IIASA model, used for the EU ozone strategy calculations, applies this approach as well.

A second point to consider is the variation of the time window during the growing season. Within the 3-month time window the daylight hours change over the season. Since this is a relatively small variation (<1.5 h) when centred around June, it is proposed to choose a uniform window over the entire growing season window, representing an average window rounded to the nearest whole hour.

The sensitivity of AOT40 to the window definition is dependent not only on the duration of the window, but also on which hours of the day contribute most to AOT40. An analysis of the sensitivity of AOT40 to the daily window was carried out for Belgium (Dumont, 1997). It was found that opening the window at 9h or even 11h CET instead of at dawn (5h) affected the AOT40 value by only a few percent, since very few exceedances of 40 ppb occur in the early morning. Because the daily ozone peak is closer to sunset than sunrise, AOT40 is more sensitive to the closing time of the window. Moving the closing hour from 21h (end of daylight) by 1 hour to 20h decreased AOT40 by 5%, and for each hour that the closing hour is moved further backward in time, AOT40 decreased by 10% (see also Figure 3.8). In an analysis for Swedish stations, a stronger sensitivity was found for stations in the Northern part, but not for Southern and Mid Sweden (see the pattern in Figure 3.9 for a station in Southern Sweden). This figure also includes data from Spain. For the Corachar and Villafranca stations, both located in mountainous surroundings, the contribution of night-time ozone is seen to be considerable. However, the sensitivity of AOT40 to the definition of daylight hours (middle part of the graph) is found to be limited. An analysis for Austria also showed the sensitivity to be relatively large for mountainous sites.

In view of the many important uncertainties in the Level I approach (see also Section 3.2.2), which is the basis for the WHO guidelines, and also because the sensitivities found are small compared with the inter-annual fluctuations of AOT40, **it is proposed to choose a fixed period for the EU as a whole, defined similarly to that used in the RAINS calculations, i.e. 8-20 h Central European Time.**

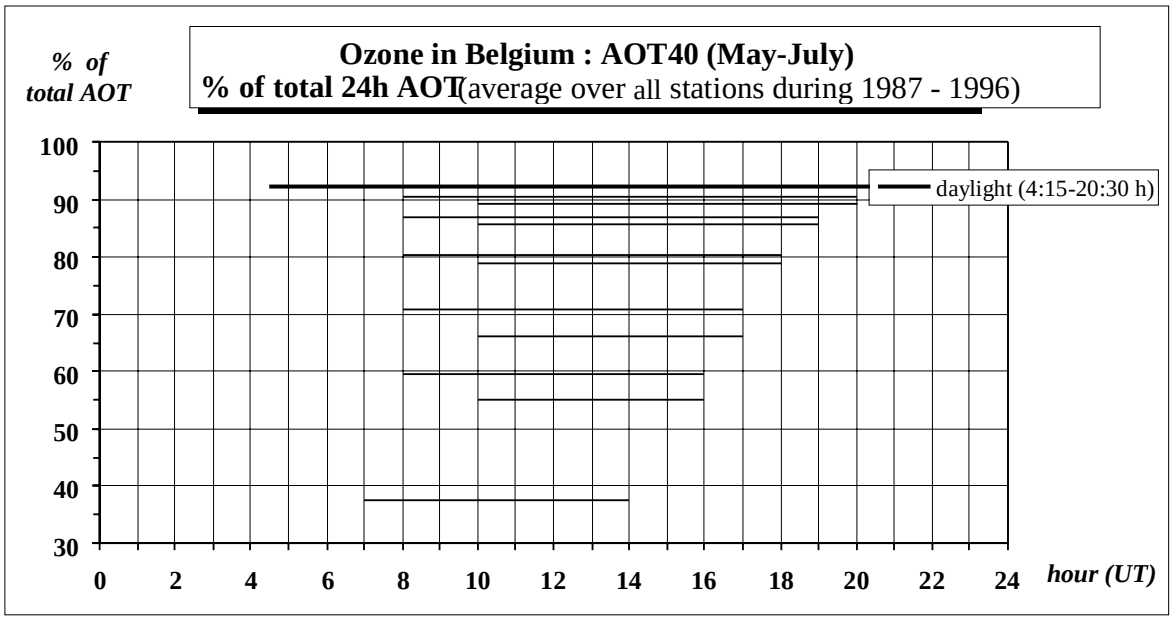


Figure 3.8. Sensitivity of AOT40 to the opening and closing hours of the daily window.

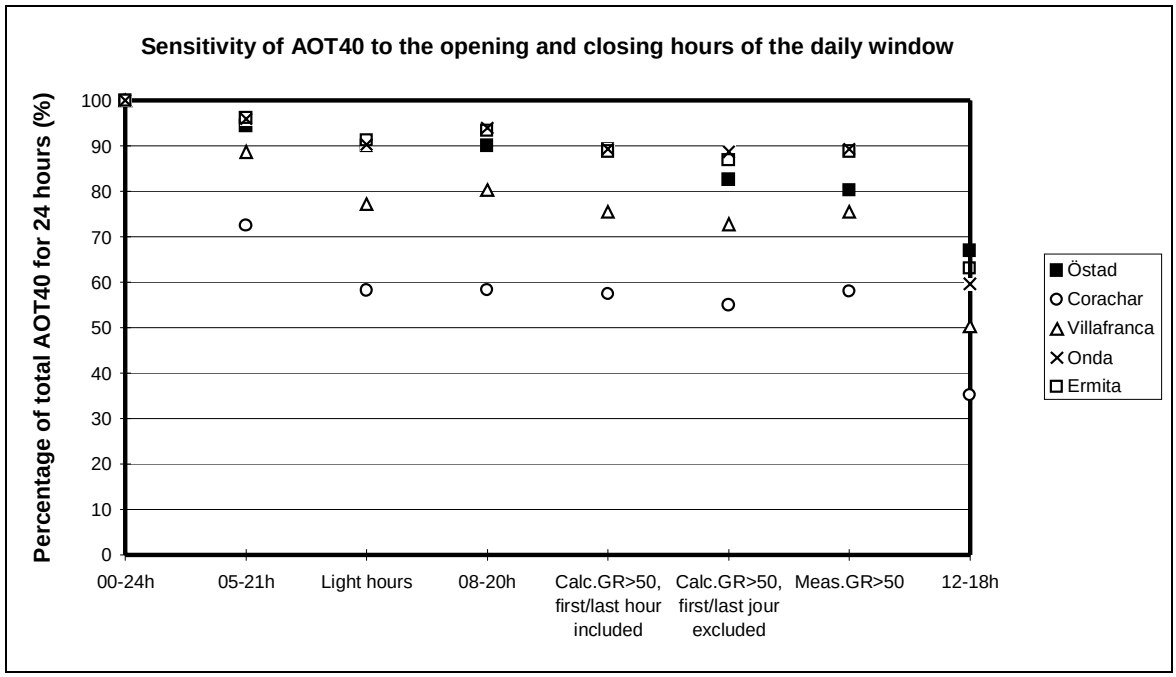


Figure 3.9. Sensitivity of AOT40 to the opening and closing hours of the daily window at the Östad station in Southern Sweden and at four stations in Valencia, Spain. GR>50: Global radiation >50 W/m^2. Global radiation is calculated for clear sky conditions. "First/last hour" refers to the whole hours in which the global radiation goes through the value 50 W/m^2.

3.7.2. Objectives for materials

The estimation of critical levels and loads is not the same for materials as for ecosystems. Since deterioration of materials is a cumulative and irreversible process which occurs even in the absence of pollutants, the concept of critical values is not applicable. Instead, threshold values are estimated based on the concept of acceptable pollution levels and deterioration rates. Whether a deterioration rate is acceptable or not depends on the specific applications and includes technical and economic considerations. Another difference worth noting is that threshold values are based on annual averages, not peak values. This is based on the assumption that dose-response relations for materials in general are linear or close to linear with respect to pollutant concentrations. For O_3 it is difficult to assign threshold levels partly because the indirect and synergistic parts have not been sufficiently quantified so far. In the UN-ECE mapping manual (UBA, 1996), however, an O_3 threshold level of 20 ppb (annual mean) was proposed for sensitive organic materials. This was based on calculations of how long it takes artists' water-colour pigments to fade in the absence of light (Shaver *et al.*, 1983) and results from exposure to ozone of paints, including automotive, industrial and household coatings, in the presence and absence of UV light (Campbell *et al.*, 1974). Another source was exposure of polypropylene in an O_3-containing atmosphere in the presence of UV radiation, which revealed accelerated embrittlement (Lucki, 1988). It should be stressed that the experimental evidence is limited and, in the two latter cases, extrapolated from accelerated laboratory exposures. The ozone level given may, nevertheless, provisionally represent a threshold above which unacceptable shortening of the material's lifetime will result (UBA, 1996). The threat which ozone poses to materials in museums, libraries and archives is reflected by the recommended limits on ozone concentrations, which indicate that they should be kept very low. Values cited in the literature, often lacking detailed justification, range between 1 and 13 ppb O_3 for some major museums or authorities in the UK, USA and Canada (Thomson, 1986) (Cass *et al.*, 1991).

The proposed level of 20 ppb is widely exceeded in the EU; 90% of the annual averages observed in the EMEP network were above it. It is proposed not to set an EU long-term objective for

materials, but to include the annual average zone concentration in the list of parameters to be reported.

3.7.3. Alert and information thresholds

Importance of information thresholds

As mentioned in chapter 2 above, it is proposed to retain the approach used in Directive 92/72/EEC of two levels of thresholds triggering the release of health-related advice to the people concerned. Today, the ozone levels at which relevant effects are known to occur are lower than at the start of the nineties, when the current Directive was developed. Consequently, it would be hard to justify ending the active dissemination of such information to the public. It could be argued that real-time information on ozone levels via the Internet is available in a growing number of Member States, allowing the people concerned to inform themselves (this will be discussed more extensively in a subsequent chapter). However, the availability of real-time information differs between Member States, and passive dissemination to the general population via the Internet is far less effective than the current system of actively informing people affected by exceedance of these thresholds. Because of this, it is proposed to retain the information system of the current Directive, although the numerical value of the levels should be reconsidered in the light of new scientific information on health effects.

The Framework Directive provides only one level, the alert threshold, for the purpose of issuing health-related information to the public. To comply as far as possible with the Framework Directive and to be consistent with other daughter directives, a two-level information system of this nature ought to be based on the alert threshold definition in the Framework Directive. It was proposed in Chapter 2 that an "information threshold" be set, defined as an alert threshold for the sensitive population, while additionally a "general alert threshold" would be set for advising the whole population.

Averaging time

Averaging times need to be defined before the levels of the information thresholds are chosen. In the current Directive the thresholds refer to 1-hour averages, but the averaging times in the WHO guidelines have been changed from one to eight hours. An 8-hour period is too long for triggering an information release, especially as the daily ozone peak typically occurs in the afternoon. A shorter averaging time is therefore preferred. For practical reasons it is proposed to use 1-hour averages for the information thresholds.

It should be noted that even with a 1-hour threshold the public will receive the information on measured exceedance *after* its occurrence, and often late in the day. It is therefore recommended that in addition to measurements of ozone levels, Member States also use a forecast system and disseminate the expected daily maximum 1-hour and 8-hour averages in the case of high ozone levels (see also Chapter 6). Such forecast systems exist in some Member States, but the current state of the art is not well enough developed to formally prescribe the application of a forecasting methodology in the Directive.

Relation between 1-hour and 8-hour maximal

Since the basis for the information thresholds is health effects related to 8-hour average ozone concentrations, a conversion ratio between the daily maximum 8-hour and 1-hour means would be useful. It should be noted that the conversion ratio varies in time and space. The variation is partly random, but systematic variations in time and space are also to be expected: at stations where the diurnal concentration pattern is relatively flat, the difference between the daily maximum of the 1-hour and the 8-hour averages is expected to be rather small. In the Netherlands the daily maximum

8-hour mean was found to be typically 80-90% of the 1-hour mean. In data provided by Austria this percentage was found to range between 77 and 96%. Low values were observed within or near agglomerations, and high values at Alpine sites. German data from Berlin indicate that the percentage is typically closer to 90% at almost all stations that are not directly influenced by traffic.

Level of the information threshold

In view of the revised WHO guideline, the levels of the information thresholds should also be reconsidered. Obviously the WHO guideline for health, 120 µg/m^3 (8-hour mean), is a possible starting point for choosing the thresholds. If a 1-hour equivalent of this (around 140 µg/m^3) were chosen for the information threshold, the number of exceedances would become much larger than under the existing Directive, and consequently in many Member States the information value would be lost. The WHO exposure-effect table (Table 3.1 in Section 3.2.1) gives a more differentiated basis for choosing the information threshold, relating the ozone level to the percentage of the population affected: the most sensitive 10% of young adults and children experience a 10% decrease in FEV_1 at 160 µg/m^3 (8-hour mean). This is also the level of the new USA standard. In ambient air 160 µg/m^3 (8-hour mean) is roughly equivalent to 180-200 (1-hour mean), which is around the current information level. (N.B. The ratio between 1 h and 8 h values in Table 3.1 is not relevant here, since the 1 h values in that table refer to exposure limited to one single hour.)

Since the current information threshold of 180 µg/m^3 is also in this range, it is proposed to set the information threshold at this level, being representative of the level where adverse effects on sensitive populations become significant. It could be defined as follows:

Information threshold is defined as an alert threshold for the sensitive population. It means the ozone concentration value beyond which there are significant adverse health effects in particularly sensitive sections of the population in the event of short exposure.

Level of the information threshold: 180 µg/m^3 (1-hour average)

Level of the general alert threshold

The levels at which no significant effects are expected to occur are now lower than some years ago, when the existing warning threshold was set. Consequently the general alert threshold is proposed to be set below the current warning threshold. Based on recent effects data, 240 µg/m^3 (1-hour mean) is regarded as the level above which significant adverse effects occur in the general population. On days when a 1-hour maximum ozone concentration of 240 µg/m^3 is reached, the following health effects may be expected to occur in the general population:

- a 50% increase in exacerbation of symptoms among adults and asthmatics,
- a 20% increase in hospital admissions for respiratory conditions,
- a decrease in pulmonary function of some 5%,
- an increase in inflammatory changes in the lung.

In healthy persons engaged for some time in physical exercise outdoors during the afternoon the following health effects may be expected to occur:

- a 15% decrease in pulmonary function,
- a 3-fold increase in pulmonary inflammatory changes.

Exceedances of the 240 µg/m^3 level have not been reported in the framework of the current ozone Directive. To acquire insight in current exceedance rates, a limited inventory was carried out of exceedances of this level. The AIRBASE data showed the average number of exceedances in the

EU to be somewhat less than one per year per station in the period 1986-1995. In 70% of the data series (a data series is one year of data from one station) no exceedances occurred, while 2 or more exceedances (with a maximum of 10) occurred in 10% of the data series. One should be aware that the representativeness of AIRBASE is limited. Some Member States provided information on exceedances observed in their networks. In Ireland and Norway no exceedances were reported in the period since 1990. In Sweden in this period exceedance was observed at one station on one day. Belgium reported that the number of days per year for which at least one site in the country registered an exceedance varied between 0 and 10 in the period 1990-1997. An analysis of data in Spain showed that the number of days on which an exceedance was observed anywhere in the country varied between 1 and 41 in the period 1990-1996. The number of exceedances per region was typically much smaller, indicating that exceedances at different stations often do not occur on the same days. This is important for judging the number of exceedances to be expected for individual zones or regions, which may be considerably smaller in size than a country, and consequently may have considerably fewer exceedance days per year than the country as a whole.

The following general alert threshold is proposed:

General alert threshold means the ozone concentration value beyond which there are significant adverse health effects in the general population in the event of short exposure.

Level of the general alert threshold: 240 $\mu g/m^3$ (1-hour average)

Information to be disseminated

Chapter 6 considers the information that should be given to the media when an information threshold is exceeded. The Member States should further be encouraged to make real-time ozone concentration data available to the public throughout the year via the Internet, Teletext or other means. This also applies to forecasts of 1-hour and 8-hour ozone concentrations for the following day(s).

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4. MONITORING AND ASSESSMENT STRATEGY

4.1. Introduction

The Directive on Ambient Air Quality Assessment and Management ('Framework Directive') contains general criteria and prescriptions for air quality assessment, to be detailed for each pollutant in the relevant Daughter Directive. Concentrations must be assessed over the whole territory of the Member States. Before a Directive enters into force, a preliminary assessment has to be made to determine concentration distributions over the territories to enable the Member States to define appropriate monitoring networks and other assessment techniques.

Several assessment techniques may be used, subject to minimum requirements regarding the number of measuring points and the type of technique; the Framework Directive links these requirements to the ratio between concentration and limit values. Since no limit values are proposed for ozone, the Framework Directive leaves assessment requirements more open in this case. Chapter 2 includes proposals to adapt the legal framework to the special case of ozone.

4.2. Monitoring and assessment under the current Directive 92/72/EEC

4.2.1. The Council Directive on air pollution by ozone

Member States currently have to comply with Council Directive 92/72/EEC on air pollution by ozone, which was adopted in September 1992. The Directive defines a number of threshold levels and establishes a harmonised procedure for monitoring and for exchanging data. It also arranges to provide the public with information when warning and information threshold levels are exceeded. Table 1.1 in Chapter 1 sets out the current threshold values.

The Directive requires Member States to forward to the Commission:

- The maximum, median and 98-percentile values of 1h- and 8h-mean concentrations;
- The number, date and duration of periods during which the information and warning thresholds shown in Table 1.1 were exceeded;
- The maximum concentration recorded during an exceedance episode;
- Exceedances of the health and vegetation thresholds.

Member States should base their information on validated continuous measurements. The data should be transmitted to the Commission within 6 months after the end of each calendar year. However, exceedances of the information and warning levels should be reported on a monthly basis. Information on siting and measurement techniques is also to be supplied. In turn the Commission prepares, via the ETC-AQ, an annual and a 'summer' report summarising all statistics and exceedances provided by the Member States (e.g. De Leeuw and van Zantvoort, 1996; Sluyter and van Zantvoort, 1996).

4.2.2. Monitoring requirements

The Directive states that complete knowledge of ozone pollution is required in all Member States and that it is necessary to use measurement stations to provide data on ambient ozone. In each Member State the ozone network is supposed to satisfy two siting criteria:

1. Stations should be located at geographically and climatologically representative sites where:

    - The risk of threshold values being approached or exceeded is highest;
    - It is likely that either the population or vegetation is exposed.

2. Additional observatories should be available in order to:

    - Help identify and describe the formation and transport of ozone and its precursors;
    - Monitor changes in ozone concentrations in areas affected by background pollution.

At this second category of stations, measurements of NO_x are mandatory and those of VOCs are recommended. Observations of NO_x and VOCs should be carried out in order to provide information on ozone formation, or to monitor transboundary fluxes of VOCs and make it possible to identify links between the different pollutants.

The Directive gives no quantitative requirements regarding network density.

4.2.3. Assessment of exposure, risk and effects in relation to network design

The basic objective of ozone monitoring under the Directive is to assess:

- The individual risk of exposure of human beings to values in excess of the health protection thresholds;
- The exposure of vegetation (forests, natural ecosystems, crops, horticulture) to the thresholds listed in Table 1.1.

However, the Directive does not describe methods of assessing exposure, risk or the effects on those at risk. For any assessment, a detailed network and site description will provide important information for evaluating the representativeness of an ozone network and for judging the air quality data from that network correctly. In its communication to the Member States on the transfer of the 1996 data, the Commission asked that they use the site classification listed in Tables 4.1 and 4.2.

Table 4.1 Basic site classification in rural, urban and street stations, used in the 1996 data reports from the Member States.

Type of station	Description
Rural	Station is located outside built-up areas of cities or conurbations. It is used to monitor 'regional background' air pollution levels. Station is located away from emission sources.
Urban	Station used to monitor 'average' air pollution levels in urban area (urban background concentration) resulting from incoming transport and from emissions in the city itself. The station is not directly influenced by emission sources such as traffic or industry.
Street	Station used to monitor air pollution levels in traffic street with more than 2000 vehicles/day within 50 meters. It is directly and predominantly influenced by traffic emissions. The station is located in an urban area but can also be operated beside a motorway.

Table 4.2 Description of immediate (0-100 m radius) and local (100 m to a few km) environment of stations. Used in transfer of 1996 data from Member States

Description of Immediate Environment within a radius of 0 to 100 m	Description of Local Environment within a radius of 100 m to a few km
□ large street heavy traffic	□ urban commercial
□ large street medium traffic	□ urban industrial
□ large street light traffic	□ urban residential
□ large street pedestrian zone	□ mixture of commercial, industrial and residential
□ small street heavy traffic	
□ small street medium traffic	□ industrial heavy concentration
□ small street light traffic	□ industrial medium concentration
□ small street pedestrian zone	□ industrial light concentration
□ canyon street heavy traffic	□ road traffic heavy
□ canyon street medium traffic	□ road traffic medium
□ canyon street light traffic	□ road traffic light
□ canyon street pedestrian zone	□ commercial
□ footway	□ residential (isolated houses)
□ front of building	□ harbour
□ terrace, bell tower, belfry	□ airport
□ courtyard, school, hospital	□ park, forest, natural field
□ trees	□ agricultural area
□ large flat area	□ mountain, valleys
□ channel	□ sea side or lake side
□ meadow, field	□ other
□ other	

4.2.4. Experiences with assessment under the current Directive

Table 4.3 indicates the number of sites for which the Member States transmitted data in their 1995 measurements. The table also gives an overview of information attached to the data reports.

Table 4.3 Numerical overview of the parameters and number of sites from which 1995 ozone data were reported by the Member States (Source: de Leeuw and van Zantvoort, 1996)

Parameter	Number of Member States	Number of sites
□ statistics/percentile values	15	844
□ number of exceedances of threshold values	15	812
□ geographical coordinates of sites	15	855
□ surroundings: information on local environment	14	745
□ information on measurement method	15	831
□ information on calibration method	14	720
□ type of site (rural, urban, street, other)	14	725
□ altitude above sea level	15	815
□ town where site is located	15	832
NO_x statistics	6	97
NO_2 statistics	7	124
NMVOC statistics	2	11

From these data, (1) the assessment and mapping of regions with (un)acceptable air quality, (2) the evaluation of effects of ozone and (3) the determination of source-receptor relations all centre around a basic question: "Does the network sufficiently cover the areas of high and typical air pollution exposure of the population and vegetation?" From the 1994 and '95 data sets reported under the Directive we conclude that the network's spatial coverage needs to be improved in many countries (de Leeuw *et al.*, 1995; de Leeuw and van Zantvoort, 1996). Blank areas are apparent in both urban and rural monitoring. The current subset of rural/background stations is estimated to cover no more than 20-40% of forests and 30-50% of crops, depending on the chosen radius of spatial representativeness of the observatories. The subset of urban/street sites covers no more than 12% of all EU15 residents. These conclusions do not necessarily imply a call for "100%" coverage of all people, crops and ecosystems by monitoring networks in the EU15. They do, however, emphasise that knowledge of the current stations' representativeness of their specific local environment needs to be improved. In each country, this could be supplemented by information on the number and extent of locations where a similar situation and air quality occurs.

4.3. The Air Quality Framework Directive and other relevant conventions

4.3.1. Assessment methods for ozone according to the Air Quality Framework Directive

In Section 2.3 it was proposed that the Daughter Directive include a long-term objective for ozone, corresponding to the critical levels defined by WHO, in combination with a Target Value, which would serve as an interim objective. No Limit Value would be set.

The Target Value should be achieved as quickly as possible within a given attainment period, whereas the long-term objective is considered an effect-based strategic reference point for the abatement strategy.

The Framework Directive links the different assessment methods to the concept of "Limit value" and the Upper Assessment Level (UAL) and Lower Assessment Level (LAL) which are defined relative to that Limit Value. Since no Limit Value will be set for ozone, the Target Value could be taken as an alternative criterion. However, since the Target Value is set higher than the Long-term Objective, inadequate assessment would be made of the extent to which the Long-term Objective is still exceeded. It is therefore proposed that the Long-term Objective alone be used as a trigger for the assessment obligation.

Study of the 8-hour averaged ozone concentrations over the past 10 years in the stations of the APIS database - supplemented by data from Finland and Austria – shows considerable interannual variability, especially in Central and Southern Europe. If UAL and LAL values for the health protection Long-Term Objective were determined in the conventional way - as in other daughter directives - they would turn out to be very low: at 60% and 50% of the health Long-Term-Objective for UAL and LAL respectively (Dumont, 1997).

Preliminary investigation of interannual variation over 10 years in some Belgian stations in the 3-month AOT40 value to protect vegetation would suggest even lower UAL and LAL values: around 25% to 35 % only of the Long-Term-Objective for vegetation.

As no Limit Value will be set and as the interannual variability of the health and vegetation related Long-Term Objectives is very high (resulting in very low hypothetical UAL and LAL), no UAL or LAL values will be defined at all.

Articles 5 and 6 deal with assessment of ambient air quality. Assessment is made in different zones in the Member States.

- The Framework Directive allows three methods: measurements, modelling and objective estimation techniques.
- Basically two types of assessment zone are defined: “agglomerations” and other (non-agglomeration) zones.

Depending on current (or initially assessed or historical) ozone levels and depending also on the type of zone, the following ozone assessment methods (or combinations) can be defined:

- Measurement is mandatory in

  1) agglomerations: zones with a population of more than 250 000 inhabitants or, where the population concentration is 250 000 inhabitants or less, a population density per km² which for the Member State justifies the need for ambient air quality to be assessed and managed.

  2) zones where levels exceed the long-term objective over a representative period.

  In accordance with Article 6(2) of the Framework Directive, when measurement is mandatory, it “may be supplemented by modelling techniques to provide an adequate level of information on ambient air quality”.

- Modelling or objective estimation techniques may be used on their own in non-agglomeration zones where the levels “over a representative period” are equal to or below the long-term objective.

  However, it also seems sensible to maintain a reduced density of monitoring stations (see 4.3.4) in non-agglomeration zones where the long-term objective is attained. These monitoring data are a valuable element of air quality assessment and complement the measurements still mandatory in adjacent agglomeration zones, in order to e.g. record the development of the large-scale ozone background and to verify and calibrate photochemical models.

The representative period referred to above should be 5 years. Relaxing the assessment requirement would be possible only if no exceedance of the Long-term Objective has occurred during the preceding five years of measurements.

The proposed ozone assessment methods are summarised in Table 4.4.

Table 4.4 Ambient air quality assessment methods according to ozone levels and type of zone

Current (or historical) ozone level	**agglomeration**	**other zone**
> LT-O	MEAS (+MOD)	MEAS (+ MOD)
< LT-O	MEAS (+MOD)	MOD or ESTT (+MEAS)

LT-O : Long-term Objective
MEAS : measurement
MOD : modelling
ESTT : objective estimation techniques

Objective estimation techniques

The term "objective estimation techniques" refers to simple mathematical methods of calculating concentrations from concentrations measured at other locations and/or times, and based on scientific knowledge of the concentration distribution. One example is linear interpolation, provided the concentration pattern is sufficiently smooth. Another example is a dispersion model that has been adjusted to reproduce concentrations measured within its domain.

4.3.2. Exchange of information (EoI)

Council Decision 97/101/EC on Exchange of Information emphasises that data collection, exchange and harmonisation of monitoring information is expected to support the Commission in combating air pollution. More specifically: EoI deals with reporting requirements, the so-called reciprocal exchange, of air pollution data. It does not set monitoring requirements. The reciprocal exchange of information covers 37 pollutants, including ozone, and incorporates a station classification scheme based on station type and location. The observation stations covered are: those under the current air quality Directives, background stations and sites operational under the previous Exchange of Information Decision (82/459). The Commission is responsible for implementation of the reciprocal exchange, though it turns to the European Environment Agency for practical implementation of EoI. Member States have to submit data on any given year by October of the following year. The datasets reported on ozone consist of the mean, median, 98-percentile, and maximum for both 1h- and 8h-averages. For some stations a selection of these statistics is reported. Furthermore, continuous 1h-average concentrations are deposited in the data archive. Until 1995 the data were stored in a database, and made available to Member States via the so-called APIS PC-application. Currently, EoI data are stored in the AIRBASE database hosted by EEA's European Topic Centre of Air Quality. This archive includes the meta-information previously stored in the GIRAFE base. AIRBASE is accessible via Internet (http://www.etcaq.rivm.nl/airbase/index.html). Currently, hourly ozone data from 50 ozone monitoring sites from 7 Member States are deposited in AIRBASE.

4.3.3. Monitoring requirements and assessment under international conventions

This section summarises ozone monitoring requirements and assessments under International Conventions to which the European Community is a Party. Each section lists the number of monitoring stations involved. N.B. Many stations may operate under several of the programmes in parallel.

Convention on Long-range Transboundary Air Pollution

The UN-ECE Convention on LRTAP hosts the Cooperative Programme for Monitoring and Evaluation of the Long-range Transmission of Air Pollutants in Europe (EMEP). EMEP provides a monitoring programme in relation to internationally agreed control measures, the so-called Protocols. The EU Member States and the European Community are Parties to the Convention.

The purpose of the EMEP monitoring network is to provide:

- Information on the spatial distribution of air pollutants over Europe;
- A basis for comparison with model calculations;
- A basis for evaluation of time trends.

The monitoring programme encompasses many compounds, including O_3, NO_2 and VOCs. Measurements of NO_2 are taken on a 24-h basis, while hourly averages are required for ozone. The pilot programme on the measurement of VOCs takes samples with variable frequency. The EMEP network is supposed to provide information with a spatial resolution of about 200 km. We note

that the LRTAP Convention is geared towards combating air pollution in rural areas. This is expressed by the programme's proposed siting criteria, which define a minimum distance between emission sources and stations of e.g. >100 m for domestic heating and >50 km for large sources of pollution (e.g. power plants). In the case of ozone this implies that the EMEP network may be of great value in assessing exposure of crops and ecosystems. It will, however, be less significant in the evaluation of human health issues. About 57 monitoring sites, including 7 measuring VOCs, in the EU15 Member States are currently connected to the EMEP photo-oxidants database. 90% data completeness is considered a realistic objective (for any details about the EMEP-network and access to data visit EMEP's website http://www.emep.int/).

Global Conventions

Relevant Global Conventions are the United Nations Framework Convention on Climate Change and the Vienna Convention for the Protection of the Ozone Layer, with its Montreal Protocol and amendments. These Conventions do not contain binding articles on monitoring of air pollutants. The World Meteorological Organisation (WMO), however, has been operating the Global Atmosphere Watch (GAW) database since 1989. This base was set up primarily to detect changes in atmospheric concentrations of greenhouse gases. This data archive includes observations of 38 ground-based O_3 sites and 15 NO_x stations in the EU15 Member States.

The WHO-GEMS/AIR programme is a global programme for urban air quality management. In the participating countries it was recommended that at least 3 stations be operated in each monitored city centre, to represent commercial, industrial and residential areas respectively. The GEMS/AIR network currently encompasses 17 cities in the 15 EU Member States.

International Air Pollution Assessments

Analyses and risk evaluations of monitoring data derived through the above Directives, Conventions and programmes have been presented in several international assessment reports, e.g. the Dobris assessment (Stanners and Bourdeau, 1995) and Concern for Europe's Tomorrow (WHO, 1995c). The authors generally used the data:

- To describe air quality in Europe;
- To compare air quality in various countries;
- To produce population/exposure relations and exposure/effect relations;
- To estimate health effects;
- To quantify physical damage;
- To study possibilities for cost-effective abatement.

These assessments concluded that the spatial and temporal coverage of ozone data in some countries in Europe is insufficient or of uncertain quality. In particular, systematic data on photo-oxidants in the urban situation is often lacking. This hampers assessment of health effects from measured data. In many cases modelled data were used.

4.4. Network design and siting criteria

4.4.1. Objectives of the network

The assessment requirements imposed by the Framework Directive mean that data need to be collected not only on ozone, but also on precursors and meteorology. This chapter therefore needs to address measurements of all those parameters. However, before describing the various measurements, we should first discuss which types of measurement should be harmonised and defined in detail by the Ozone Directive.

The measurements should be specified in relation to the purposes of the assessment. In the Framework Directive the following main purposes can be distinguished:

1. Compliance checking;
2. Air quality management and understanding ozone;
3. Public information.

a. Compliance checking

A major purpose of monitoring is to check whether ambient ozone levels lie within the various air quality thresholds (Target Value, Long-Term Objective, Alert Threshold, Information Threshold). This means not only that ozone concentrations as such should be monitored, but also that the representativeness of the measurements should be clearly known. In the case of ozone, the conversion of ozone into NO_2 due to titration by local NO has proved to be the main factor determining representativeness. Because of this, NO_2 measurements will be considered an inherent constituent of ozone measurement for compliance checking. *Compliance checking, which may trigger compulsory action to reduce emissions, needs to be uniformly defined, and so the Ozone Directive should prescribe ozone and collocated (at the same location) NO_2 measurements in detail.*

b. Air quality management, understanding ozone

To manage air quality, the authorities need to know the sources of ozone. Measurements of ozone by itself are of very limited use in determining the sources causing ozone formation. Monitoring to understand and manage ozone should include concentrations of precursors, in particular NO_x and VOCs, and to some extent CO. Meteorological data are also essential for relating ambient ozone levels to sources. *Over-detailed prescriptions for measuring precursors and meteorological parameters would often hamper rather than assist Member States' efforts to perform optimal air quality management. So these measurements should not be specified in detail.*

c. Public information

An important goal of monitoring is to inform the public on air quality with respect to ozone. For clarity, this should focus on ozone only. NO_2 and other precursor measurements performed in relation to ozone are often not characteristic for exposure to these pollutants, therefore public information on those substances should not be defined by the ozone directive. *Measurements for public information should be directly related to the legal thresholds and should also be harmonised within the EU. These measurements should therefore be specified in detail.*

It should be noted that the two purposes of NO_x measurements entail two different measurement strategies: NO_2 measurements related to ozone compliance checking require that ozone and NO_2 be measured at exactly the same location, whereas there is no need to perform precursor measurements at ozone monitoring sites for air quality management purposes. Further, only NO_2, and not NO_x, is relevant for compliance checking, while NO_x measurement (usually including NO_2 and NO separately) is important for air quality management.

So, detailed prescriptions will be given in this document for ozone and collocated NO_2 measurements. Measurements of precursors and meteorological parameters will be recommended in more general terms; guidance will be given regarding the number and location of measuring sites. Table 4.5 summarises purposes and related monitoring requirements for the various parameters.

Forecasting of ozone should also be mentioned here as a purpose of monitoring. Its purpose relates both to air quality management and to public information. Since no single, generally accepted methodology exists, measuring for this purpose should not be specified in detail.

Table 4.5 Purposes/requirements of monitoring pollutants and meteorological parameters

	Ozone	*NO_2*	*NO_x*	*VOC*	*CO*	*Meteo*
Purpose of monitoring						
a. Compliance checking ozone levels	✓	✓				
b. Air quality management, understanding ozone	✓	✓	✓	✓	✓	✓
c. Public information on ozone	✓					
Requirements	++	++	+	+	+	+

++ Detailed prescription in the ozone directive
+ General recommendations in the ozone directive

4.4.2. Network design

The Framework Directive requires a preliminary assessment to be carried out in order for data to be available by the time the legislation is implemented. These data are used to select an assessment regime in accordance with the Framework Directive. In practice, most Member States already have an ozone monitoring network. This can be used to conduct the preliminary assessment; if necessary, it may be complemented by other techniques.

An ozone monitoring network can be designed by performing the two following basic steps:

1. Estimation of the spatial and temporal distribution of ozone concentrations (preliminary assessment)

2. Formulation and implementation of a measurement strategy (siting criteria).

The first step means determining, as far as is technically possible, where the areas of exceedance are and when concentrations of concern occur. As stated in the Framework Directive, this preliminary assessment of ozone concentrations should also take into account information regarding the localisation of possible receptors such as population and sensitive vegetation. The complexity of this task will depend on the particular features of each Member State, which may mean that different techniques and methods need to be applied in each case. Below are some of the most common information and techniques to help perform this task:

- Geographical information: orography, land use, population distribution, etc.;
- Meteorological and climatological information, meteorological surveys;
- Previous measurements, indicative measurements, bio-indicators;
- Emission inventories, mathematical models.

The second basic step is to devise and implement a measuring strategy such as will meet the objectives of the network. A crucial aspect of this strategy, which has a great impact on the final design of the network, will be the siting criteria for monitoring stations. This issue will be discussed in the following sections.

The flow chart below summarises the process described above.

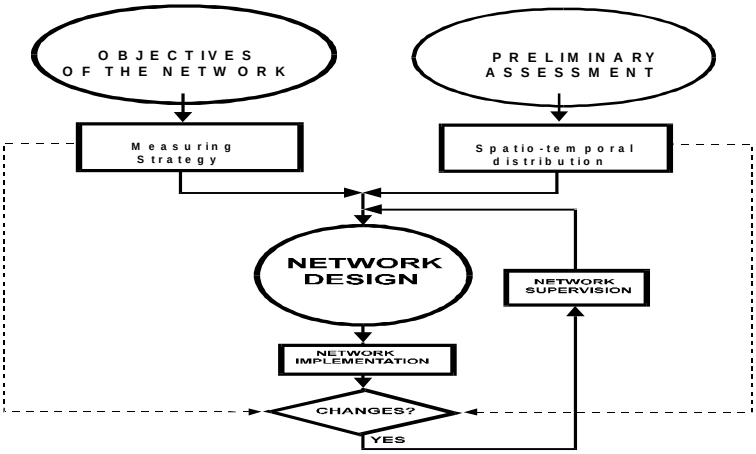

Figure 4.1. Network design flow diagram

In designing an ozone monitoring network, it must be kept in mind that the spatial and temporal distribution of ozone concentrations is not the same as for other pollutants. This should have a direct impact on network design. This issue is discussed in the following section.

4.4.2.1. General implications of spatial and temporal distribution of ozone levels for network design

Monitoring design will depend primarily on the spatial distribution of ozone and of human population and vegetation, for which air quality objectives are set. In principle, monitoring should be conducted where areas of exceedance are expected and where the relevant receptors are present. Thus network design should be mainly influenced by the (very different) spatial distribution of both population and vegetation.

Even at small scales, ozone levels can produce a very heterogeneous pattern, especially in complex terrain. In contrast to primarily emitted pollutants such as PM, NO_x or SO_2, the highest ozone concentrations are usually observed not in city centres or at kerbside, but in areas away from sources of ozone precursors, such as suburban and rural areas. The largest contribution from photochemical ozone formation comes downwind of conurbations, usually in a plume anything from 20 up to 150 km downwind, which is where the highest short-term concentrations can usually be observed.

Knowledge of the spatial and temporal distribution of ozone, mainly with respect to areas of exceedance/no exceedance, is the basis of network design. This is especially true for complex, mountainous terrain, where ozone concentrations largely depend on altitude.

Ozone has a specific distribution in mountain areas, where high concentrations are fairly constant due to greatly reduced dry deposition, insignificant depletion by NO and enhanced exchange processes with the reservoir layer; high long-term values (i.e. 8-hour mean value) are usually significantly higher at higher altitudes. (Loibl W. et al, 1994)

Ozone concentrations show: very marked seasonal variation, with maximum levels during spring and summer; annual variation, especially in those zones where meteorology plays an important role in ozone levels (southern Europe); and also diurnal variation.

Ozone concentrations show diurnal variations that depend on the combined effects of:

- Photochemical ozone formation;
- Horizontal and vertical ozone transport;
- Ozone depletion processes by dry deposition;
- Ozone depletion by NO titration.

It should be kept in mind that ozone measurements for compliance analysis should be carried out only in areas relevant for the specific receptors (population and vegetation). In mountainous areas, measurements conducted in valleys and basins are not representative for ozone levels in higher lying areas.

4.4.2.2. Network optimisation

Network optimisation means making best use of available resources to meet the objectives of the network. Periodical review of networks is needed because of:

- Possible changes in the network's objectives (new legislation/requirements, etc.);
- Availability of better knowledge of the spatial/temporal distribution of ozone patterns (new measurements, better knowledge of ozone phenomenology, enhanced assessment/monitoring techniques, etc.);
- Long-term changes in concentration levels and patterns.

A number of techniques are available for network optimisation. Techniques using statistical criteria are very useful for:

- Identifying areas where additional monitoring sites should be installed;
- Identifying 'redundant' monitoring sites in order to thin out the monitoring network.

4.4.3. Siting criteria

One of the main objectives of the Framework Directive is to harmonise the monitoring networks of the various Member States in order that air quality may be assessed in a uniform manner throughout the EU. The complexity and variety of the EU Member States makes the definition of common siting criteria a key element for accomplishing this task. If ozone concentrations are to be assessed reliably (which includes determining areas where thresholds are met/exceeded, and trends in these areas), good station representativeness and sufficient spatial coverage are of crucial importance.

4.4.3.1. Classification of stations

Stations could be classified according to what levels are measured, (i.e. ratio O_3/Ox), the objectives of the measurement (protection of health/vegetation, research), or the surrounding environment of the station. For network design purposes, it has been found most practical to classify monitoring stations according to surrounding environment.

Classifying a station in a particular category can be difficult. There are different ways of doing it:

1) It can be assessed, just by looking at its surroundings, whether a monitoring site is urban, rural or something else (though with spurious conclusions in some cases),

2) Through appropriate handling of station data (i.e. ratio O_3/Ox, with Ox = O_3 + NO_2), it can be possible to find out whether it is e.g. a suburban or rural/regional or rural/background site (in this case, the station may be said to tell "by itself" how it should be classified).

A study has been carried out on data (monthly average of 1-h concentration data) produced by collocated NO_x and O_3 instruments in various areas of France. A first finding of this study was that sites could be placed in the following three categories:

- In urban areas, O_3/Ox ratios are fairly low all year round: 0.1 in winter and 0.5 in summer. This means that the oxidant in sampled air is mostly composed of nitrogen dioxide at those sites (urban/background sites).
- At rural/regional sites, O_3/Ox ratios range widely, from 0.2 in winter to 0.95 in summer. The oxidant burden of the air is composed mainly of ozone in summer, but not in winter due to the influence of city emissions.
- At rural/background sites, conversely, O_3/Ox ratios are higher, and close to 1 all year round (>0.80). This means that the oxidant in sampled air is mainly composed of ozone all year round.

Regarding the local station environment (i.e. human activities typical of the zone, land use, type and quantities of local emissions, etc.), the station classification that seems most appropriate would be:

Table 4.6 Monitoring site classification

TYPE	DESCRIPTION OF THE LOCAL ENVIRONMENT
TRAFFIC	Stations directly and predominantly influenced by traffic emissions and generally located in the urban area or along a motorway.
URBAN	Stations located in the densely built-up cores of cities but not directly influenced by local emission sources such as traffic or industry.
SUBURBAN	Stations located on outer fringes of cities; can also include sites in adjacent areas immediately influenced by the emissions of a larger city.
RURAL	Stations located in rural areas, which might include small settlements. These stations might be under the influence of urban or industrial plumes.
RURAL BACKGROUND	Stations located in rural areas outside built-up areas of cities or conurbations and very far away from big emission sources.
INDUSTRIAL	Stations directly influenced by industrial emissions.
SPECIFIC	Stations located in a very specific location such as on top of a communications tower or tall building, and not for compliance analysis.

A more detailed classification might be necessary for data interpretation and modelling purposes. Description of the immediate environment (radius of up to 100 m) and local environment (radius of between 100 m and a few kilometres), as defined in AIRBASE with a total of 40 codes, might be useful.

4.4.3.2. Other parameters to be measured

Not only ozone, but also ozone precursors should be monitored to obtain basic information for devising ozone reduction strategies and to perform a complete assessment. Ozone precursor data are needed :

- to verify emission inventories;
- to analyse emission trends;
- to classify ozone stations;
- to study ozone formation processes.

As stated in Section 4.3.1 above, measurement of NO_2 at the location of an ozone station gives important information on the representativeness of the ozone measurement. Accordingly, it is proposed that ozone stations also be required to measure NO_2. For reasons of efficiency, it would be useful if these collocated NO_2 measurements could also be used for the NO_2 Directive. However, the locations with the highest NO_2 levels do not coincide with the areas where ozone is highest, so the collocated NO_2 measurements are not necessarily part of the NO_2 assessment for the NO_2 Directive.

Table 4.7 shows which parameters are mandatory/recommended at each station type in the framework of the ozone Directive.

	O_3	NO_2	NO_x
Urban	XXX	XXX	X
Suburban	XXX	XXX	X
Rural	XXX	XXX	X
Rural background	XXX	XX	X
Specific	XX	XX	

Parameters to be measured per type of station:

XXX	Mandatory
XX	Recommended
X	Desirable at some stations

The table does not address obligations for other Directives. Precursor measurements are described in more detail in Section 4.6 “Ozone precursor measurements”. Measurements of meteorological parameters have not been included in Table 4.7 since the need for such measurements will depend very much on the availability of data from meteorological stations. Meteorological information is extremely useful in assessing the representativeness of ozone measurements. In mountainous terrain and/or areas with frequent land-sea breeze phenomena, *in situ* meteorological measurements, preferably wind, temperature and solar radiation, are highly recommended. In uniform terrain, meteorological parameters from a nearby weather station are often sufficient. This is often the case also within cities, where meteorological measurements are usually difficult to run and costly for a routine air quality monitoring programme. In research projects and modelling exercises, wide information on the wind pattern over the whole research area is of great value. See also Section 4.3.3.5 “Meteorological measurements”.

4.4.3.3. Macroscale siting criteria

This section details the objectives of the different monitoring station types mentioned above and some siting criteria that should be followed in order to reach those objectives.

Table 4.8: Macroscale siting criteria

TYPE OF STATION	MEASUREMENT OBJECTIVES	REPRESENT-ATIVENESS	MACROSCALE SITING CRITERIA
TRAFFIC	to validate emission inventories; to help determine the magnitude and type of precursors emitted by the city; to determine the exposure of humans and materials to precursors and other pollutants (fulfilment of other Daughter Directives.).	Street levels	in street canyons or at kerbsides within the urbanised area; near to motorways; where maximum precursor levels are expected; where representative measurements of pollutant levels can be obtained; This type of station is generally not appropriate for measuring ozone, due to its low representativeness. For NO_x measurements, the NO_2 Daughter Directive should be considered.
URBAN	to determine the ozone concentrations to which people and materials within the urban atmosphere are exposed (compliance analysis for human health); to know the well-mixed precursor concentrations present in the urban air shed; to verify photochemical models; to help in the development and verification of future reduction strategies; to perform trend analysis;	A few km^2	away from the influence of local emissions such as traffic, petrol stations, etc.; vented locations where well mixed levels can be measured; adequate locations such us residential and commercial areas of cities, parks (away from the trees), big streets or squares with little or no traffic, open areas characteristic of educational, sports or recreation facilities;

TYPE OF STATION	MEASUREMENT OBJECTIVES	REPRESENT-ATIVENESS	MACROSCALE SITING CRITERIA
SUBURBAN	to determine the exposure of the population and vegetation located in the outskirts of a big city to high ozone concentrations (compliance analysis for human health and vegetation); to verify photochemical models; to help in the development and evaluation of reduction strategies; to help towards a better understanding of ozone phenomenology; to perform trend analysis.	Some tens of km^2	at a certain distance from the area of maximum emissions, downwind following the main wind direction(s) during conditions favourable to ozone formation; where population, sensitive crops or natural ecosystems located in the outer fringe of an agglomeration are exposed to high ozone levels; other locations that might provide information about ozone phenomenology; Where appropriate some suburban stations also upwind of the area of maximum emissions, in order to determine the regional background levels of ozone and precursors present in the air before it enters the urbanised area. This would help in the assessment of ozone concentrations, the establishment of boundary conditions for the use of models, and also in episode forecasting.
RURAL	to determine the exposure of population, crops and natural ecosystems to sub-regional scale ozone concentrations (compliance with health and vegetation thresholds); to help in the assessment of sub-regional ozone concentrations; to help in the establishment of boundary conditions for the use of models; to help in the development and evaluation of reduction strategies; to help towards a better understanding of ozone phenomenology; to perform trend analysis of ozone and precursors;	Sub-regional levels (a few hundred km^2)	stations can be located in small settlements and/or in areas with natural ecosystems, forests or crops; representative for ozone away from the influence of immediate local emissions such as industrial installations and roads; in open area sites, but not on higher mountain tops;

TYPE OF STATION	MEASUREMENT OBJECTIVES	REPRESENT-ATIVENESS	MACROSCALE SITING CRITERIA
RURAL BACK-GROUND	to provide information on the exposure of crops and natural ecosystems to regional scale ozone concentrations (compliance with vegetation limit values); to provide information on regional ozone background concentrations; to help in the establishment of boundary conditions for the use of models; to help in the development and evaluation of long term reduction strategies; to help towards a better understanding of ozone phenomenology; to perform trend analysis of ozone and precursors;	Regional/national /continental levels (1 000 to 10 000 km^2)	Station located in areas with lower population density, e.g. with natural ecosystems, forests, far removed from urban and industrial areas and away from local emissions; avoid locations which are subject to locally enhanced formation of ground-near inversion conditions, also summits of higher mountains coastal sites with pronounced diurnal wind cycles of local character are not recommended; In general, EMEP siting criteria might be valid for this type of station.
INDUSTRIAL	to validate emission inventories; to assist in the determination of the type and magnitude of the precursors emitted by an industrial activity; to know the exposure of people and vegetation to ozone precursors and other pollutants.	Industrial area	located within or at the edge of the area of maximum industrial emissions; sited where a representative (spatial and temporal) measure of precursors can be obtained; This type of station is not appropriate for measuring ozone, due to its low representativeness. For NO_x measurements, the NO_2 Daughter Directive should be considered.

4.4.3.4. Microscale siting criteria

Once the macroscale location of a monitoring station has been identified, the following criteria should be observed in order for measurement of ozone levels in the station's immediate environment to be representative.

Vertical and horizontal probe placement:

- Located 1.5 to 4 metres above ground level (higher ones are possible for URBAN stations);
- More than 1 metre vertically or horizontally away from any supporting structure;
- Far from any furnace, incineration flues or air-conditioning outlets.

Spacing from obstacles:

- Distance between any obstacle and the inlet probe at least twice the height that the obstacle protrudes above the sampler;
- Unrestricted airflow in an arc of at least 270º around the inlet probe;
- If probe is located on the side of a building, 180º clearance is required.

Table 4.9 Requirements concerning distance from roads for urban, suburban and rural stations

Average daily traffic (vehicles/day)	Minimum separation distance (metres)
<10 000	> 10
15 000	20
20 000	30
40 000	50
70 000	100
>110 000	> 250

4.4.3.5. Meteorological measurements

Meteorological measurements are required for a correct assessment of ozone concentrations (short-term forecasting, modelling, interpretation of episodes, etc.). Such measurements should be representative of the meteorological conditions that affect pollutant transport and dispersion within the area to be monitored; this means the meteorological measurement does not necessarily have to be performed at the same location as the ozone measurements. Measurements should be made away from trees, buildings, steep slopes, ridges, cliffs, and hollows. Surface monitoring ($h \leq 10$ m) of wind speed/direction, temperature, relative humidity, barometric pressure and solar radiation is important, but upper air meteorological monitoring is also encouraged in certain areas/periods. Measurements for evaluation of the mixing properties of the atmosphere are also encouraged.

Where possible, it will be useful to follow WMO guidelines for siting meteorological measurements. Meteorological information might often be available from national meteorological organisations. In any event, it needs to be available within a short period of time in order to fulfil its purpose.

4.4.4. Number of ozone stations

In Chapter 2 it was proposed that minimum assessment requirements be related to the long-term objective, and not to the target value. Furthermore, the Framework Directive stipulates that in "agglomerations"[18] at least one fixed measuring station should be operated where an alert threshold has been established. It is proposed that two assessment regimes be defined:

- One for all agglomerations, and for zones where ozone levels exceeded one or both long-term objectives during a specified period;
- One for other zones, i.e. non-agglomeration zones where neither of the long-term objectives is exceeded during a specified period.

For the sake of continuity, the specified period should not be too short, but nor should the station network follow changes in ozone levels too slowly; a period of five years seems a good compromise between stability and flexibility.

In conformity with the earlier Daughter Directive, it is also proposed that a distinction be made between situations in which ozone assessment is based solely on measurement data from ozone stations, and situations in which the measurement data are generalised to build up a territory-covering picture of ozone levels using modelling or other mathematical techniques (supplementary assessment).

Table 4.10 shows the proposed minimum number of stations for agglomerations and zones where levels exceed a long-term objective and where measurement is the sole source of information. For urban, suburban and rural stations, the minimum relates to the number of inhabitants of the agglomeration or zone; for rural background stations it was considered better to express the minimum in terms of spatial coverage. The working group also discussed an alternative of setting the minimum number of stations in "other zones" depending on the area rather than on the size of the population in a given zone. Eventually it favoured the approach presented in Table 4.10.

A further alternative was brought up emerging from concerns about the different number of stations per inhabitants in urban and rural zones as proposed in Table 4.10. It suggests that the definition of the subcategories of the category "other zones" be modified on the basis of population density and depending on the share of people living in rural communities. Presented at a late stage it could not be discussed further due to time constraints.

In the cases described in the table, collocated NO_2 measurements should be conducted at at least 50% of the ozone stations.

It is proposed that a reduction be allowed in the number of stations by up to one third in the case of supplementary assessment, but with a minimum of one station in each agglomeration or zone. In this case, nitrogen dioxide should be measured at each station, except at rural background stations.

These numbers represent the minimum. However, networks should be dense enough to provide a description of spatial distribution that is sufficient to reliably assess how ozone levels develop in relation to the target values. The number of stations should, for example, be increased in the case of zones under the influence of an urban plume.

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[18] The Framework Directive defines 'agglomeration' as a zone with a population concentration in excess of 250 000 inhabitants or, where the population concentration is 250 000 inhabitants or less, a population density per km^2 which for the Member State justifies the need for ambient air quality to be assessed and managed.

For zones where levels are within the long-term objectives, the number of stations should be sufficient to confirm compliance with the long-term objective and to follow the ozone trend. To this end, supplementary assessment techniques could be used. In suburban areas of agglomerations and rural areas around agglomerations, the minimum should be one-third of the number of stations specified in Table 4.10. If the minimum number of stations calculated for a zone is zero, stations in neighbouring zones should be taken into account for the assessment. The density of rural background stations should not be below 1 per 100 000 km^2.

Table 4.10 Minimum number of ozone stations for agglomerations and for zones where levels are above the long-term objective and where measurement is the sole source of information

	Agglomerations		**Other zones**		
Population (x1000)	Urban	Suburban	Suburban	Rural	Rural background
< 250	0	0	0	1	1 station / 50 000* km^2
< 500	0	1	1	1	as an average
< 1 000	0	2	1	2	density over all zones
< 1 500	1	2	1	3	per country
< 2 000	1	3	1	4	
< 2 750	2	3	1	5	
< 3 750	2	4	1	1 add. station	
> 3 750	2	1 add. station	1	per 0.5 m inh.	
		per 2 m inh.			

* 1 station per 25 000 km^2 for complex terrain in regions below 55ºN latitude

Measurements of precursors, particularly NO_x and VOCs, have different purposes than ozone measurements, so the measuring strategy is in principle quite different. A strategy for such measurements is not being proposed here, but it is important that guidelines for the strategy and a general framework for the results be developed. This could be done during the implementation of the new ozone directive.

4.5. Methods of measuring ozone

The measurement of ozone can be divided into three separate steps:

- Sampling;
- Measurement or analysis;
- Calibration.

The following tables give the most commonly used methods and their main advantages and disadvantages.

4.5.1. Existing sampling methods

Method	Description	Reference	Advantages/Disadvantages
1. Laminar flow method	Flow 150 l/min, tube diameter 15 cm Inert material: glass, stainless steel, Teflon	EPA	+ isokinetic sampling, sample unaffected - unnecessarily large construction
2. Turbulent flow manifold	Modular sugar cane design Inert material: glass, stainless steel, Teflon		+ low cost, modular construction - loss of particulates (dirt), possible loss of O_3
3. Sampling without manifold	Direct connection of analyser inlet to station sampling head		+ low cost, efficient sampling

4.5.2. Existing measuring methods

Method	Description	Reference	Advantages/Disadvantages
1. Manual methods			+ cost-effective - discontinuous and time consuming measurements
1.1 BAKI	Absorption of ozone in boric acid buffered potassium iodide + photometry	(Flamm)	- not ozone specific
1.2 NBKI	Absorption of ozone in neutral buffered potassium iodide + photometry	EPA	- not ozone specific - overestimation
1.3 Indigo sulfonic acid method	Absorption of ozone in solution of indigo sulfonic acid + photometry	VDI 2468 Bl. 5	- possible interferences
1.4 Diffusive sampling	Diffusive sampling onto absorbent (e.g. DPE, KI, Indigo) + photometry		+ cost effective - integrated measurement over several days
2. Automated methods			+ continuous, real time measurement - requires regular calibration and maintenance
2.1 Chemilumi-nescence method	Measurement of chemi-luminescence reaction between ozone and ethylene	ISO 10313, VDI 2468 Bl. 4	- ethylene flammable gas
2.2 UV photometric method	Measurement of UV absorption by ozone	ISO CD 13964	- possible interferences (humidity, some VOCs)
3. Optical methods			+ simultaneous multi-component analysis - integrated concentration over path length
3.1 DOAS spectrometry	Differential optical absorption along path length		+ easy, maintenance-free operation - expensive analyser, measurement disturbed by fog - field calibration complicated
3.2 DIAL spectrometry	Differential optical absorption of backscattered laser beam		+ 2D-mapping possible - very expensive analyser - field calibration complicated

4.5.2.1. Measurement method performance

The most commonly used measurement method is the UV photometric method. Analyser performance is typically as stated in the Annex to ISO document CD 13964.

In line with Directive 92/72/EEC, the European Reference Laboratory of Air Pollution organised two inter-laboratory exercises to compare calibration procedures and measurements in the national laboratories. 21 laboratories from EU and EFTA countries took part in the exercises. The overall agreement of the results based on UV photometry was within +/- 5%. A comparison between the three different calibration techniques (GPT, UV reference photometer and the boric acid buffered method) showed a maximum deviation of +/- 2%. Water vapour interference in the instrument response was tested - and observed - for almost all analysers.

4.5.2.2. Indicative measurements

On-line monitoring of atmospheric pollutants in air quality monitoring networks generally requires expensive and sophisticated measurement techniques. Simpler measurement techniques, called indicative or screening techniques, may offer a cost-effective alternative to conventional techniques. The diffusive sampling technique is the most interesting of them.

A diffusive sampler consists of a tube, one end of which contains a chemical substance that fixes the pollutant. The pollutant is sampled onto the absorbent at a rate controlled by the molecular diffusion of the pollutant in the air. The amount of pollutant collected by the sampler is a function of the ambient air concentration integrated over the sampling period. After exposure of the samplers over a period of a few days, the tubes are closed and returned to the laboratory for analysis by colorimetric techniques. This sampling technique is still being developed for application to ozone.

The main advantage of the diffusive sampler is that it does not require a pump or electrical power and that it runs unattended during the sampling period. The most important drawback is that it yields a time-integrated measurement over a long period (typically one week), and that concentration peaks such as those occurring during episodes are hardly detected.

The DOAS is a useful alternative to conventional methods. Ozone and NO_2 can be measured simultaneously at preferably urban background and rural background sites.

4.5.3. Existing calibration methods

Method	Description	Reference	Advantages/Disadvantages
1. UV method	Measurement of UV absorption caused by ozone	ISO CD 13964, VDI 2468 Bl. 6	+ primary calibration method + precise and accurate
2. KI method	Bubbling in absorbing solution + photometry		+ cost effective - unknown accuracy
3. GPT/NO_2-Detection	Titration of NO with O_3, detection of NO_2 gives the amount of O_3		+ common method also for NO, NO_2 calibration

4.5.4. Reference measurement method

The reference method of the current Directive is the UV absorption method. On the basis of experience, and intercomparisons carried out by ERLAP, there is presently no need to change the reference method, so the following is proposed:

- Analysis method: UV absorption method (ISO CD 13964);
- Calibration method: Reference UV photometer (ISO CD 13964, VDI 2468, p. 6).

4.5.5. Quality Assurance and Quality Control of measurements

Quality assurance is a system of procedures to ensure that:

- Measurements are of known precision and accuracy,
- Results are comparable and traceable,
- Data are representative of ambient conditions,
- Optimal use is made of resources.

The major elements of a quality assurance programme are:

- Network design (see separate section): number of stations, siting criteria;
- Measurement technique: sampling, analytical and calibration procedure;
- Equipment evaluation and selection: validation of methods, test of instrument performances;
- Routine site operation: calibration under field conditions, maintenance, management and training.

QA/QC procedures are described in the WHO UNEP GEMS/AIR Methodology Review Handbook Series, Volume 1, "Quality Assurance in Urban Air Quality Monitoring".

At present, QA/QC programmes only exist in a few monitoring networks in the EU Member States and with varying degrees of efficiency. A European intercomparison of calibration procedures for ozone measurements with 21 laboratories from 12 Member States has shown good agreement between results. A field intercomparison regarding the quality of routine measurements in networks is still required for ozone.

With monitoring networks due to expand with the implementation of the framework Directive, a lot of new laboratories, including many private companies, are expected to be in charge of monitoring. This will require particular measures to ensure the quality of measurements and the capability of laboratories:

- Accreditation of laboratories: various standardised QA/QC systems have been developed in recent years, such as the Good Laboratory Practice (OECD), the ISO 9000 and the EN 45000 laboratory accreditation procedures. The EN 45001 procedure was developed by CEN in collaboration with the Commission and is the best adapted for testing laboratories in the field of air pollution measurements. Laboratories seeking accreditation are audited by a national or international accreditation organisation. This audit mainly concerns aspects such as laboratory installation and equipment, qualification and training of personnel, proper quality control, technical audit and traceability of measurements. Applications for laboratory accreditation are the only enforceable way of ensuring an effective QA/QC procedure.
- Validation of measurement methods and standardisation at CEN or ISO level.
- Certification of equipment: test of instrument performance (the development of a standardised CEN test procedure is therefore urgently needed).
- Organisation of intercomparison at EU level: organisation by the Commission of EU wide intercomparison exercises (round robin tests, inter-laboratory exercises, spot checks in the monitoring networks) to ensure international comparability of measurements.
- Publication by the Commission of guidance documents, organisation of training and workshops.

4.6. Modelling Requirements for the Ozone Directive

4.6.1. Objectives

Modelling makes two key contributions to air quality assessment:

- Optimised station siting

  It is essential to devise a system in which a relatively limited number of monitoring stations, supported by other assessment techniques, can be sited to ***represent air quality*** (ozone and precursors) reliably throughout a Member State.

- Generalisation of measurements

In zones where concentrations *show many potential areas of exceedance*, e.g. in an agglomeration or where there are many small towns or villages within a larger zone, stations at one location could represent similar locations elsewhere. This is referred to as generalisation.

Measurements only provide information on temporal variations in ozone concentrations. To assess the effects of ozone on human health and the environment, information is also required on the spatial distribution of ozone in order to show potential areas of ozone exceedance. For this generalisation, detailed photochemical transport models or merely interpolation schemes could be applied, where justified.

Moreover, air-quality modelling enables short and long term forecasts of ozone concentration to be carried out for public information. The planning of short-term measures can be analysed and the relationship of air quality to emissions established in a quantitative sense. Furthermore, air-quality modelling provides a better basis for describing areas of exceedance in the entire zone and serves to create the essential insight into the mechanism of ozone formation and for emission reduction scenario calculations. It also provides (not only additional) essential information for the management of air quality in the zone, as required under the Framework Directive.

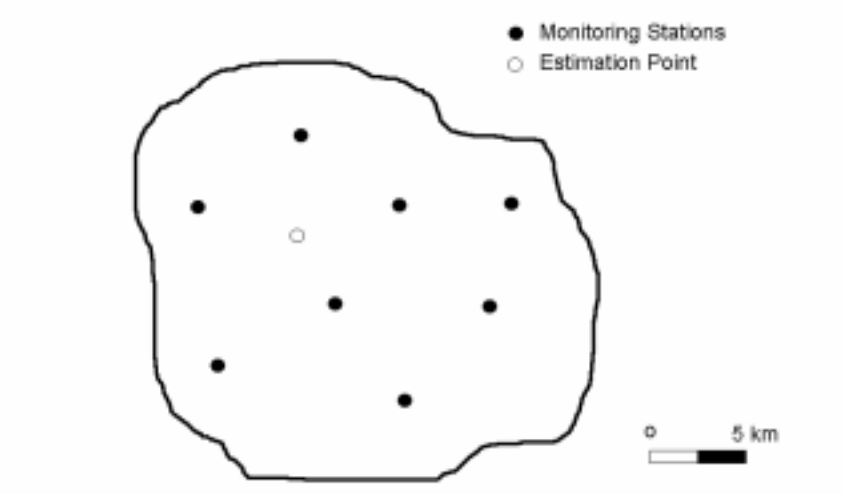
4.6.2. Key features of models for photochemical transport

An ambient air quality model is a means whereby pollutant emissions can be related to atmospheric pollutant concentrations. For ozone, the relationship with precursors is non-linear, and these ozone models are essential for predicting the emission reductions necessary to achieve a desired Air-Quality Standard.

Ambient air quality models can be divided, broadly speaking, into three types: **empirical models, interpolation models** for the generalisation of measurements and **analytical physical reality models**.

The **empirical models** are based on statistical analysis of past air monitoring data with the aim of generalising with suitable extrapolation/interpolation techniques. Large urban air sheds contain a number of air monitoring stations operated under the auspices of air pollution control authorities, at which 15-min to 1-hr average pollutant concentration levels are reported. A great deal of information is potentially available from these enormous databases, and the statistical analysis of air quality data.

The limitations of this type of model are that despite the accuracy at the measuring site, estimations at other points of the domain are subject to interpolations. The uncertainty of these interpolations can be reduced by establishing a dense network or by suitably varying the positions of the measuring sites. In either case the associated costs and time spent establishing reliable time series of the air quality measurements at the new site are high. These air-quality measurements can to some extent be linked to emission sources, provided meteorological data are available, to demonstrate how emission changes affect air quality concentrations.



A more advanced category of empirical model is based on interpolation schemes coupled with measurement data as the basis for temporal and spatial assessment of ozone levels. Numerous **interpolation models** have been used to calculate rural ozone levels, especially for the determination of AOT40 values (Schneider *et al*., 1996, Gimeno *et al*., 1996, Lövblad *et al*.,1996). These models have a different degree of sophistication. Either the measured values are directly interpolated or specific modifications are taken into account. Factors which can be considered in interpolation schemes are height above mean sea level and relative height. The diurnal variation of ozone levels can largely depend on an area's altitude.

The advantage of interpolation models is that they rely on measured data. Consequently, the models are really a supplement to the measurements. However, it is very important that the stations be sited to be most representative, which means this kind of model should only be applied to areas where extensive monitoring has been carried out. Additionally, in certain northern European domains some of these models can be applied in real time so that the information on actual spatial distribution can be evaluated immediately.

Analytical physical reality models simulate the atmospheric processes affecting pollutants by means of atmospheric transport and chemical processes. The model will involve considerations of emission patterns, meteorology, chemical transformations and removal processes. Because of the inherently random character of atmospheric motions, one can never predict with certainty the distribution of concentration of marked particles emitted from a source. Although the basic equations describing turbulent diffusion are available, there is no single mathematical model suitable for computing atmospheric concentrations over all ranges of conditions.

Analytical models use the Eulerian or Lagrangian approach to consider atmospheric dispersion .

In Eulerian-type models, the atmospheric region of interest (several thousands of km²) is subdivided into a three-dimensional grid and this framework is fixed in space. Pollutants enter into and are transported out of each grid cell while at the same time the material and chemical balance is calculated for each grid cell. A formulation of this type is very useful because Eulerian statistics are readily measurable and because the mathematical expressions are directly applicable to situations in which chemical reactions are taking place. Each calculation is repeated at each time step, giving a new concentration pattern for the entire domain of interest. Eulerian models provide a good means of predicting three-dimensional concentration distributions over a region. They do require significant calculating times, though these have fortunately been drastically reduced thanks to the improvements in computer speed and multiprocessor architectures.

The Lagrangian class of model describes the pollutant content of a moving parcel of air according to the displacements of groups of particles released in the fluid. During its course, the air parcel

receives emitted material from the sources it passes over, chemical reactions occur, and the pollutant content is calculated at selected receptor points. The mathematics of this approach are more tractable than those of the Eulerian methods, but the applicability of the resulting equations is limited because of the difficulty of accurately determining the required particle statistics. Though this type of model is less demanding computationally, air quality is only calculated along the trajectory and we require many trajectories to describe regional air pollution.

Under certain idealised conditions the mean concentration of a species emitted from a point source has a Gaussian distribution. This fact, although strictly true only in the case of stationary, homogeneous turbulence, serves as the basis for a large class of atmospheric **plume models,** which are in common use for stacks or individual line sources. For ozone, models of this type can idealise the mean concentration downwind of an urban centre (more than 100 km) if this urban area is considered a continuous, elevated point source. In which case the resolution of the origin and the scale of emissions at the source will only be rough approximations.

The latest generation of tools is based on **integrated assessment modelling** simulations. These take into account the effects of multi-pollutants in the environment and the relative cost/benefit of different control strategies. In order to do so, several modules describing the emissions, control costs, (photo)chemical and physical processes are linked together, making it possible to try out control strategies and find cost-effective solutions. The EU ozone strategy was developed using a model of this type, the RAINS model (see Chapter 5). Such "models" are in principle suitable for real time visualisations as well as for examining the environmental impact of certain technical or non-technical scenarios. Their uses include:

1. Establishment of emission control legislation.
2. Evaluation of proposed emission control techniques and strategies.
3. Planning of locations of future sources of air pollutants.
4. Planning for the control of air pollution episodes.
5. Assessment of responsibility for existing levels of air pollution.

4.6.2.1. The Modelling Strategy

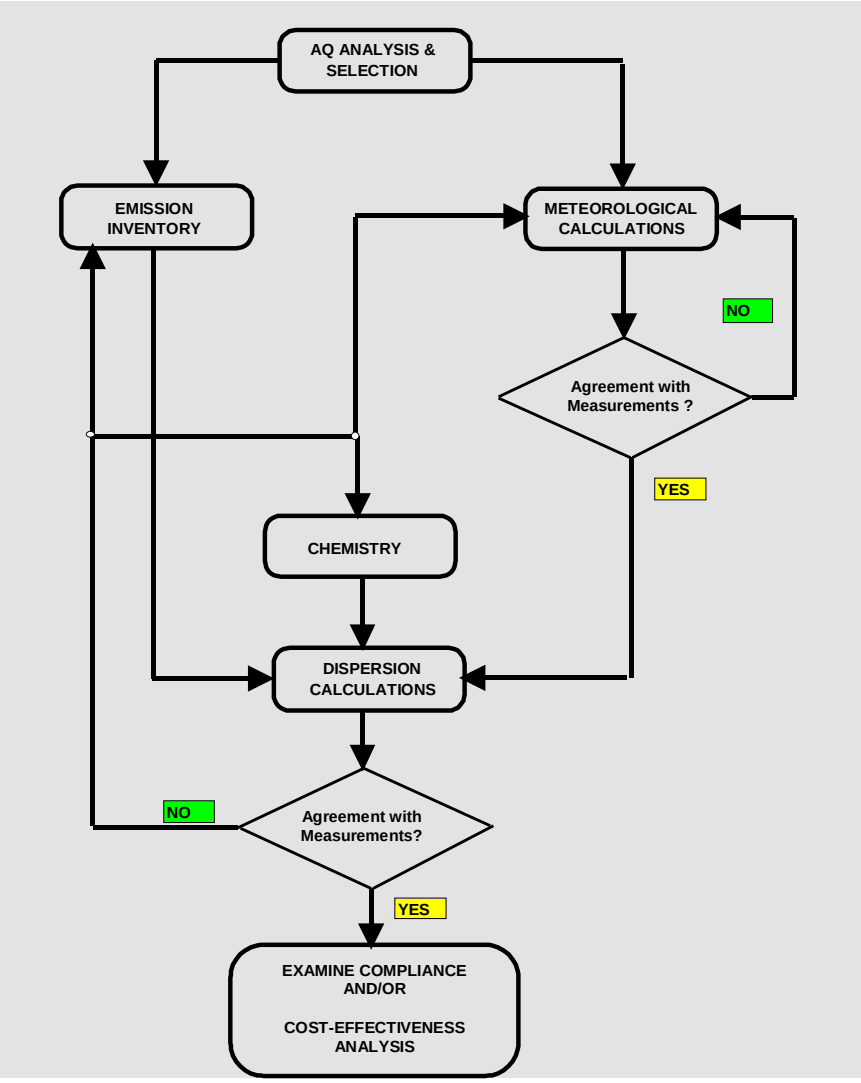
For almost all modelling approaches, the strategy employed contains the elements illustrated in the flowchart below.

An initial picture of air quality concentrations within an area is essential both for identifying the representative period over which modelling will be conducted and for fixing calibration points against which the success of the modelling simulation will be compared.

The meteorological analysis will establish regular fields necessary for the dispersion calculations.

The chemical scheme must be made consistent with the emission inventory to account for the appropriate chemical species. Species are not of equal importance in North and South European domains.

Following the successful completion of the dispersion calculations, the relationship between air concentrations and emissions can be made explicitly. With this relationship, cost data can be associated in order to identify the best scenario for cost-effectiveness or maximum cost benefit.



4.6.2.2. Preliminary considerations

The primary consideration before conducting any modelling assessment is which model to select. This paper does not wish to create the impression that this decision could be simplified or even automated. None of the photochemical models could be taken off the shelf, and in any case the process of adaptation to domain requires the experience of a specialist for a number of issues such as:

- Overall size and resolution of the domain.
- Complexity of the terrain.
- Precursors and the accuracy of emissions rate.
- Physical processes.
- Removal mechanisms (deposition).
- Chemical scheme.
- Type of meteorological model (diagnostic, prognostic, assimilation).
- Boundary and initial conditions.
- Time step and duration of the simulations.
- Output requirements and the recording frequency of concentrations.
- Validation criteria, etc.
- Setting simulation objectives and procedures for checking spatial or temporal compliance.

Different pollutants pose different needs relative to the spatial scales of modelling. For example, carbon monoxide poses a local problem in the vicinity of busy roads and intersections. Thus, to

assess the effectiveness of motor vehicle emission controls on CO levels requires a model with spatial resolution the order of the width of a city street. On the other hand, photochemical ozone is usually a region-wide problem, caused by area-wide emissions of hydrocarbons and oxides of nitrogen. This is not necessarily the case for domains with many city agglomerations, where transport of ozone might become significant.

4.6.2.3. Spatial and Temporal Considerations

The spatial resolution of an ambient air quality model (i.e. the area over which the predicted concentrations are averaged) may range from several metres to several thousand kilometres.

The partial differential equation(s) are normally solved numerically by a process that requires the continuous concentration field to be approximated by a discreet grid of points. The choice of the spatial grid on which the equations are solved is governed by the degree of spatial detail in the emissions inventory and the meteorological variables available. With a spatial resolution of 2 km, the spatial resolution of the predicted concentrations can be no smaller than 2 km.

Model	**Size of Domain**	**Typical Spatial Resolution**
Micro Scale	200 m x 200 m x 100 m	5 m
Urban Scale	100 km x 100 km x 5 km	2 km
Regional Scale	1000 km x 1000 km x 10 km	20 km
Continental Scale	3000 km x 3000 km x 20 km	80 km

It is sometimes useful to predict pollutant concentrations in the immediate vicinity of sources, i.e. the city centre as well as the greater downwind area. In such cases, the spatial resolution of the concentrations might be as small as a few km at the centre but coarser at the outer regions. The coupling of these resolutions in multi-nested layers and the on-line interaction of the solution schemes (two-way nesting) provides the basis for accurately examining man-made emissions and their impact in greater geographical areas.

Apart from the improved treatment of boundary conditions with nested simulations, another obvious advantage of this type of modelling is that simulations can be run for coarser domains over long modelling periods (several months).

The temporal resolution of an ambient air quality model (i.e. the time period over which the predicted concentrations are averaged) may vary from several minutes to one year. For example, a model may predict the 15-min average pollutant concentration as a function of location. The requirements in implementing a model will be strongly governed by its temporal resolution.

4.6.2.4. Interpretation of Modelling Results

Concentrations calculated from photochemical simulations could be expressed in terms of:

Contour plots: presentation of concentration fields and the spatial max. and min. values

Time series: for calculating exceedances, over episodes or annual mean conditions

Tables: appropriate for presenting concentration statistics

Temporal Frequency Distributions: assessing the temporal characterisation of air quality and the accumulated percentiles obtained from a fixed monitoring station.

Spatial Frequency Distributions: assessing the spatial variability of air quality and the identification of hot spots over the monitoring domain.

Accumulated Exposures: hours of persistent high concentrations. Suitable for assessing exposures and crop damage over a certain time period.

4.6.2.5. Uncertainty versus Consistency

The deciding factor in judging the validity of a theory for atmospheric diffusion is the comparison of its predictions with experimental data. It must be kept in mind, however, that the modelling results are based on predicting the ensemble mean concentration <c>, whereas a single experimental observation constitutes only one sample from the hypothetically infinite ensemble of observations from that identical experiment. Thus, it is not to be expected that any one realisation should agree precisely with the predicted mean concentration even if the theory used is applicable to the set of conditions under which the experiment has been carried out.

In evaluating the air-quality models the following list of statistical parameters can be utilised:

$< \overline{C}^{t}_{obs} >$: Mean value of observations
$< \overline{C}^{t}_{pre} >$: Mean value of predictions
σ_0 : Standard deviation of observations
σ_p : Standard deviation of predictions
NMD : Normalised mean difference
RMSE : Root mean square error
$RMSE_s$: Systematic RMSE
$RMSE_u$: Unsystematic RMSE
a, b : Intercept and slope of regression line
Corr. : Correlation coefficient
IA : Index of agreement

Where, t denotes the temporal variation for t = 1,...,T and the subscripts ‘obs’ and ‘pre’ refer to observed and predicted values respectively. In most circumstances these statistical parameters do not indicate the uncertainty of the results. However, the magnitude of the divergences has no physical significance and the calculated values have no absolute meaning. It is therefore proposed that only the comparisons of the mean value and the standard deviation be maintained.

For ozone modelling it is more important to have a consistent tool over the large physical domains than to be accurate at a single point and be able to deviate orders of magnitude by interpolations at intermediate positions. For this reason validation of the simulations is far more important than statistical uncertainty at a fixed position.

4.6.2.6. Validation

A model based on statistical analysis of pass air monitoring data might predict concentrations at one or more stations (essentially “point measurements”) or the average of the readings at a number of stations, intended to represent average pollutant concentrations in the region. In a model based on the solution of continuity equations for each pollutant, the concentrations are essentially concentrations averaged over a cell with a predefined size, and thus represent volume-averaged values.

The aim is to compare results from the simulations against data collected over the domain from all monitoring stations. It is useful to establish confidence levels against monitoring data, to examine the reliability of the models and to test the consistency of the modelling features implemented.

The objectives of validation tests are: to avoid unrealistic modelling circulation, to identify wrongly initiated parameters, to realistically simulate daytime and night-time differences and to test the temporal and spatial accuracy against time-series of monitoring data.

The following list of criteria are proposed:

- Comparison of mean measured and calculated values for over as many stations as possible or for the integral over the whole city.
- Comparison with measurements of diurnal air-quality concentrations for monitoring stations for which data are available.
- Compare the mean, max. and standard deviation of calculated values against measurements.
- Compare the exact location of the maximum concentrations over the domain.
- Compare the mean, max. concentration at the city centre with measurements for daytime and night-time. The location of the maximum concentrations over the domain should be also considered.
- Compare the time-series of the wind-fields (direction and speed) generated by the atmospheric models on the hourly basis.
- Comparison of the temporal frequency distribution of hourly concentrations for the modelling period. Examine the same frequency from the annual database of hourly measurements.
- Examine the spatial frequency distributions over the modelling domain.
- Examine the above for several pollutants involved in photochemical simulations.

4.6.3. Short-Term Forecasting

In the current ozone Directive, Article 5 requires Member States to take the necessary steps for the public to be informed if certain thresholds are exceeded. This information includes forecasts of changes in concentration, in the geographical area concerned, and in duration.

The goals of ozone episode information and forecast are, explicitly or implicitly (van Aalst et al, 1996):

- To satisfy public information requirements;
- To further reduce and prevent exposure;
- To warn the authorities, industry and the public to take emission reduction measures;
- To increase public support for structural measures.

In general, these goals require that reliable information is provided in good time. Typically, measured information has to be available in near real time, and forecasts are needed at least one day in advance.

Development of air pollution forecasting has a long history, and started at least 20 years ago (see e.g. Revlett, 1978). A recent worldwide review of currently operational systems has been prepared by AIRPARIF (Fromage, 1996). Operational forecast systems can be found in Europe, the USA, Canada, Japan, Hong Kong, Mexico, South Korea, and Australia; in Europe outside the EU, Switzerland operates such a system.

Experience and developments in 10 EU countries have been recently reviewed by a Technical Working Group on Data exchange and Forecasting for Ozone episodes in Northwest Europe (TWG-DFO). The group was convened by the European Environment Agency at the request of

environment Ministers at a conference in London in May 1996. Experts from Austria, Belgium, Denmark, Finland, France, Germany, Luxembourg, the Netherlands, Sweden and the United Kingdom participated, under the coordination of the European Topic Centre on Air Quality. The results are available in a report (TWG-DFO, 1997).

The report reviews systems for forecasting and information on ozone episodes, as currently in use and under development. The systems operate on the basis of (combinations of):

- Empirical methods and/or expert opinions;
- Statistical models;
- Causal models.

In statistical models, the prediction is generated from real-time measured ozone and meteorological data, combined with statistical information on the most likely evolution of the concentration in the given or predicted meteorological conditions. The statistical information is extracted from a database of measurements over several years. In so-called artificial neural networks, the system is self-learning in the sense that it optimises predictions on the basis of earlier experience.

In causal models, the concentration is calculated from emissions of ozone precursors (volatile organic compounds and nitrogen oxides) and forecast meteorological conditions in Europe, taking into account relevant atmospheric processes such as dispersion, transport, chemical conversion and deposition. Key information for causal models are up-to-date emission inventories of ozone precursors, reflecting recent implementation of emission reduction measures (Skouloudis, 1997, 1998).

The report also discusses and proposes criteria for testing the validity and reliability of forecast systems, and recommendations for evaluation procedures are provided. It is recommended that testing be carried out for several summers, and a range of skill parameters used. In the information presented to the public, forecast uncertainty should be indicated in simple language. The Working Group recommends that countries evaluate their forecasting systems using the proposed procedures, and discuss the results at a workshop.

Experience in European countries has shown that expert judgement on the basis of additional information can increase the performance of more formal ozone episode prediction systems considerably. Information from measurements in countries upwind may improve the quality of the forecast.

The report comes with a recommendation for a simple data exchange system on the Internet. Seven European countries now routinely exchange very recent or current ozone concentration data on a pilot system operated by AEA Technology (UK). (Stedman,1998)

Several European countries, both EU Member States and central European countries, have, in the past few years, exchanged information on air pollution information and forecasting systems in a series of informal workshops on international exchange of data for smog warning and air pollution information systems in Europe.

The TWG-DFO and the EEA propose to extend the activities in collaboration with southern and central and eastern European countries.

4.7. Ozone precursor measurements

4.7.1. Requirements of the current directive 92/72/EEC

The current Directive recommends measuring precursors of ozone in selected stations of the air quality monitoring networks. However, the Directive does not specify:

- What the objectives are of monitoring the precursors;
- Which precursors are to be measured besides nitrogen oxides;
- In which network stations these precursors should be measured;
- Which potential measurement techniques should be used.

These questions therefore need to be addressed with priority, before advice can be given on the measurement of ozone precursors.

4.7.2. State of implementation in the EU networks

While the recommendation to measure nitrogen oxides was followed in all the Member States (as part of the NO_2 directive), only a few of them have monitored hydrocarbons properly. Only in two countries (UK and NL) was the measurement of VOCs extended to the national air quality monitoring network. In some other Member States (B, F, DK, D, I, S) these measurements were confined to regional networks and generally consisted of non-automated techniques (sampling on adsorbent cartridges) or automated BTX measurements.

It should further be noted that background levels of O_3 precursors (hydrocarbons from C_2 to C_8 and some carbonyls) have been monitored by the EMEP network since 1992-93. These precursors are measured with a view to developing and validating a photochemical pollution model. The list of precursors is very similar to the list proposed by EU experts presented in 4.6.4. The measurements are performed regularly in about 10 network stations, by grab sampling on canisters, every Monday and Thursday at 12.00 h.

4.7.3. Monitoring objectives and measuring strategy

Measurement of ozone precursors can support various possible monitoring objectives, such as:

- Surveillance of precursor emissions, in order to check the efficiency of emission reduction strategies, to assess emission trends and as a tool for source attribution. It should however be noted that the use of a single monitoring station is not appropriate for assessing emissions from a stationary source, in particular if emitted by a high stack. It can be used in urban areas where about 70% of the precursors are emitted by traffic, in order to track emission trends and evaluate the efficiency of abatement measures.

- Research purposes, supporting the understanding of ozone formation mechanisms and the elaboration/application of models. A full understanding of the complex ozone phenomenology would, however, require much more information, such as on other photo-reactive species, and in particular detailed meteorological data. The organisation of intensive measurement campaigns during photo-chemical episodes therefore appears to be a more effective tool to support this objective.

- Public information. Except for some species such as benzene, most VOC precursors do not represent an acute and direct risk for human health. Even if the precursors are at the origin of ozone formation, the mechanism is mainly controlled by meteorology. As a secondary pollutant, ozone is generally not produced where the precursors are emitted. For these reasons, this objective is not considered very relevant.

Surveillance of precursor emissions from traffic in urban areas is therefore the only relevant objective for measuring precursors on a regular basis in the monitoring networks. Short-term measurement campaigns are considered more effective for assessing emissions from elevated single sources, as well as biogenic emissions.

4.7.4. Selection of precursors

Both nitrogen oxides (NO and NO_2) and volatile hydrocarbons are known to be the major precursors of ozone formation.

NO_x

For precursor measurements, NO_x is more relevant than NO_2. However, it is important to know the fraction of NO_2 in NO_x emissions, since directly emitted NO_2 contributes differently, more directly, to ozone levels than NO.

VOCs

The incidence of a single ozone VOC species on the formation of ozone depends directly on its chemical reactivity (also called the photochemical ozone creation potential – POCP) and on its concentration in ambient air. Based on these two parameters, a selection of the most relevant VOC precursors for ozone formation was established in 1990 by the EC Working Group on VOC Measurement Techniques of the Photochemical Pollution Steering Committee. This resulted in a list of 32 VOC precursors, including hydrocarbons from C_1 to C_9, as well as formaldehyde and carbon monoxide (see table 4.11). This list is very similar to EMEP's list of most relevant VOC precursors.

Table 4.11: List of the most relevant VOC precursors established by the Working Group "VOC Measurement Techniques"

Methane	1-Butene	Isoprene	Ethyl benzene
Ethane	trans-2-Butene	n-Hexane	m+p-Xylene
Ethylene	cis-2-Butene	i-Hexene	o-Xylene
Acetylene	1.3-Butadiene	n-Heptane	1.2.4-Trimeth. benz.
Propane	n-Pentane	n-Octane	1.2.3-Trimeth. benz.
Propene	i-Pentane	i-Octane	1.3.5-Trimeth. benz.
n-Butane	1-Pentene	Benzene	Formaldehyde
i-Butane	2-Pentene	Toluene	Carbon monoxide

From two intercomparison exercises of VOC measurements organised by ERLAP in 1991 and 1994, it has appeared that:

– Not all the EU Member States are equipped to perform the full list of VOC precursor measurements at the level of the air quality monitoring networks;

– The quality of the measurements is not sufficient to allow a sensitive analysis of the results as required to comply with the monitoring objective;

– The high cost of performing the full list of VOC precursor measurements cannot be justified, particularly in view of the poor resulting data quality.

Although measurement of the full list of precursors is considered the most valuable approach, the complexity of the task and its cost make it mostly unpractical. Alternatively, it is possible to reduce the list of precursors to a limited number of hydrocarbons representative of different VOC emission categories, and to use simpler measurement techniques:

– Measurement of BTX, or carbon monoxide, or TNMHC (total non-methane hydrocarbons), as representative species of automotive emissions. It should however be noted that emissions of BTX, CO and TNMHC depend greatly on the composition of gasolines and the characteristics of the car fleet. These parameters may show strong variation from one Member State to another, and may even fluctuate within one Member State. The use of these pollutants as an indicator for precursor emissions therefore requires regular calibration with respect to a full hydrocarbon analysis by gas chromatography;

– Measurement of particular hydrocarbons, as representative species of industrial emissions.

Measurement of the full list of hydrocarbons or of their indicators is not mandatory but recommended. It is, however, recommended that the full list of ozone precursors be measured during measurement campaigns.

4.7.5. Selection of stations - siting criteria

It is important not to confuse the objectives and measuring strategies of various types of measurements. The objectives of precursor measurements only partly coincide with the objectives of ozone measurements, so the associated measuring strategies are also different. Locations close to traffic and industrial plants that emit ozone precursors (refineries, petrol storage, thermal power plants without denox filter) may be suitable for investigating precursor emissions, but are not suitable for characterising ozone levels. Consequently ozone measurements are not always useful. The purpose of mandatory NO_2 measurements collocated with ozone is to characterise the representativeness of the ozone stations (Sections 4.3.1 and 4.3.3.2), so the strategy for NO_2 measurements should be regarded as separate from the strategy for ozone precursors. In addition, measurement of NO_2 in the framework of the NO_2 Daughter Directive should be regarded as separate from NO_x precursor measurements, since NO_2 measurements aim primarily at checking compliance of NO_2 levels with the Daughter Directive. This caveat is not meant to imply that each objective should have a separate network, but one should realise that in many cases the station siting criteria are not compatible.

Table 4.12 gives an indication of relevant locations for VOC measurements.

Table 4.12: Measurement of VOC indicators

Type of station	
Traffic	XX
Urban	X
Suburban	XX
Rural	X
Rural Background	X
Industrial	XX

XXX: Mandatory
XX: Recommended
X: Desirable at some stations

Measurement is recommended at traffic, suburban and industrial sites. It is desirable at urban, rural and rural background stations. It is attempted here to develop a measuring strategy for precursor measurements. This should be done in a different framework.

4.7.6. Measurement techniques

Measurements of NO_x, CO and Benzene (Toluene, Xylene) should be performed in accordance with the recommended measurement techniques specified in the respective AQ directives (currently being prepared).

The sum of total volatile organic compounds – after separation of methane (TNMHC) – is normally measured by flame ionisation measurement. It is regularly calibrated by reference gas mixtures of a single component (propane, butane). The results are expressed as the amount of carbon per volume.

These measurements should be calibrated regularly by performing an analysis of the full hydrocarbon composition (fingerprint).

During measurement campaigns, the full list of VOC precursors is to be measured using the gas chromatographic method.

4.8. Data quality objectives

Data quality objectives must be established to comply with assessment objectives. They will be defined in terms of required accuracy (trueness and precision), minimum time coverage and minimum data capture. Below, these requirements are expressed as the expected precision of the assessment methods. For the time being, the potential for linking the requirements directly to the assessment regime is not considered.

Required accuracy for O_3, NO and NO_2 measurement (expressed as maximum uncertainty for averaging time at the limit value):

- Continuous measurements: +/- 15% (individual measurements)

- Indicative measurements: +/- 30% (individual measurements)

The proposed values are based on the performances that can be achieved by implementing techniques corresponding to the current state of the art for the various methods, and taking into account the provisions of Article 3 of the Framework Directive (approval of measuring devices ensuring accuracy of the measurements, quality assurance programmes organised by the Commission).

Accuracy of measurement is defined as laid down in the "Guide to the Expression of Uncertainty of Measurements (ISO 1993), or in ISO 5725-1 "Accuracy (trueness and precision) of measurement methods and results" (1994). The percentages in the table are given for individual measurements, averaged over the period considered by the limit value, for a 95% confidence interval (bias + two times the standard deviation).

The accuracy for modelling and objective estimation is defined as the maximum deviation of the measured and calculated concentration levels, over the period considered by the limit value, without taking into account the time scale of the events.

Time coverage of the measurements:

- Automatic analysers: 100% (continuous or quasi-continuous)
- Indicative measurements: > 10% (during the summer period)

Minimum data capture (availability of valid data):

- Continuous measurements: 90% during summer, 75% during winter.

The requirements for minimum data capture and time coverage do not include losses of data due to the regular calibration or normal maintenance of the instrumentation.

Validity Criteria:

A minimum of 75% of the individual 1h measurements should be available for an 8h value to be considered valid. The same criteria is proposed for the AOT40, related to the respective time window. In order to obtain a valid 8h daily maximum value from a series of hourly running 8h values, 75% of the 8h averages between 6 and 21 hours CET should be present.

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(2) ETC-AQ: http://www.etcaq.rivm.nl

(3) EMEP: http://www.emep.int

5. ABATEMENT STRATEGY AND COST IMPLICATIONS

5.1. A cost-effective EU-wide strategy

5.1.1. Introduction

Council Directive 92/72/EEC required the European Commission to 'submit to the Council a report (Beck *et al.*, 1999) on the evaluation of photochemical pollution in the Community'. This report was to be 'accompanied by any proposals the Commission deems appropriate on the control of air pollution by ozone and, if necessary, for reducing emission of ozone precursors'. As described in Chapter 1, the Air Quality Steering Group and various associated working groups, together with other bodies, assisted the Commission in this work.

Chapter 1 gives an overview of observed ozone trends. After a continual increase in ozone levels in Europe over the last century or so, the general trend over the last decade has been unclear. It is not possible to extrapolate from past trends to predict future ozone levels because emission reductions of ozone precursors in past and future years will alter the trends. We must instead rely on model predictions to estimate future ozone levels.

It is uncertain how global emission changes will affect hemispheric background levels. Emissions in Asia for example are expected to increase significantly. On the other hand, and probably more importantly, emissions from Europe and North America are expected to decline. For the USA, emissions are expected to fall from 21 million tonnes VOCs and 23.8 million tonnes NO_x in 1990 to 15.3 million tonnes VOCs and 21 million tonnes NO_x in 2010 (EPA, 1997). Additional reductions for 22 Eastern States, currently under discussion, would reduce the 2010 NO_x emissions by a further 1.2 million tonnes. In the scenario analysis described below, the background levels were assumed to remain unchanged between 1990 and 2010.

The RAINS model (Amann *et al.*, 1998) was used to investigate the ozone situation in 2010, anticipated on the basis of existing and planned national and international policies in Europe - the so-called 'Reference Scenario'. These calculations showed that, compared with 1990, a decrease in ozone levels is to be anticipated almost everywhere in Europe, particularly regarding the higher concentrations in summer episodes. Indices for average EU population exposure above the WHO guideline were predicted to decrease by about 63% while the exposure to vegetation above the guideline for crops would fall by 41%.

Existing and planned emission reduction measures will lower ozone levels in the EU significantly, but they will not solve the ozone problem in large parts of Europe. Additional measures are needed, but even then it is clear that no feasible strategies exist to attain the long-term ozone objectives in the foreseeable future. Because of this, it is proposed in Chapter 2 that the strategy for the next decade be based on an interim target value, to be set at a level attainable by 2010. This approach establishes a direct link between the setting of the target value and the development of the ozone strategy, which in turn is closely linked to other developments, in particular the EU acidification strategy and the development of the multi-pollutant protocol in the framework of the UN-ECE Convention on Long Range Transboundary Air Pollution, which aims at reducing acidification, eutrophication and tropospheric ozone. Because of these links, it was decided to integrate all these developments and to base the various strategies on one common technical basis. In terms of EU legislation, the National Emission Ceilings Directive and the Ozone Daughter Directive constitute direct legal implementations of these strategies.

The wide array of environmental goals and the complex manner in which exceedances of environmental targets depend on emissions of SO_2, NO_x, VOCs and NH_3 made it necessary to rely on computer modelling to identify cost-effective ways of improvement. The model used in this work was RAINS.

5.1.2. Cost optimisation methodology

The methodology of the RAINS model has been extensively described elsewhere (Amann *et al.*, 1998) and the references given therein. The model, developed at IIASA, comprises modules for emission generation (with databases on current and future economic activities, energy consumption levels, fuel characteristics, etc.), for emission control options and costs, for atmospheric dispersion and chemistry of pollutants (based on the EMEP model) and for environmental sensitivities (i.e. databases on critical loads). To create a coherent and comprehensive picture of options for simultaneously addressing the three environmental problems (acidification, eutrophication and tropospheric ozone), the model considers emissions of sulphur dioxide, nitrogen oxides, ammonia and VOCs. The model has the same spatial resolution as the EMEP model, from which the chemical dispersion module was derived, i.e. about 150 km x 150 km. This fact adds a certain degree of uncertainty to the possibilities of interpreting the results for small countries with extensions close to this grid-size level. The same limitation applies to the assessment of the ozone exposure of urban population because the model cannot resolve spatial changes in the ozone concentration on the urban scale.

For the strategy development the model was operated in its 'optimisation mode' to identify cost-optimal allocations of emission reductions to attain specified environmental targets. For ozone, two types of environmental target were imposed simultaneously, related to the 'gap closure' and 'ceiling' approaches respectively (see Chapter 2). In the gap closure approach, the 'gap' between the pollution level in 1990 and the protection level desired in the long term is quantified for each grid cell of the model, and a fixed percentage reduction ('closure') of this gap is imposed throughout the EU for 2010 as an optimisation restraint. The levels aimed at in this approach vary across the Community and depend on the base situation in 1990. The 'ceiling' approach represents the target value of the ozone Daughter Directive: it sets a uniform pollution level for each grid cell throughout the EU. The approaches, including several refinements, are described more extensively in the IIASA report (Amann *et al.*, 1998) and to IIASA's website http://www.iiasa.ac.at/~rains/.

In the optimisation calculations the gap closure and ceiling approaches were used simultaneously for both the health-related and the vegetation-related target values, and in addition a gap closure was defined for acidification. Many combinations of these five types of gap closures and ceilings were tried out to find a proper balance between improved environmental quality on the one hand and the cost of emission reductions on the other. This process of model simulations, discussions between the various EU and UN-ECE groups and IIASA, modifications and refinements led to the 'central' H1 scenario, which the majority of the Steering Group viewed as a good basis for the political discussions to follow.

The ozone levels arrived at in the RAINS model simulations could not be directly used to set the target values. Two limitations in the model had to be compensated for as far as possible:

A. Recalculation of the number of Exceedances from AOT60

Time and technical constraints precluded a restructuring of RAINS to allow optimisations for the target value for health in the form proposed in Chapter 3, i.e as number of days with exceedances above an 8h average of 120 $\mu g/m^3$. Instead, a surrogate value, AOT60[19], was used to define the optimisation target. After the optimisation a post-processing step

[19] AOT60 was defined in analogy to AOT40: the accumulated exposure above the threshold concentration of 60 ppb (120 $\mu g/m^3$) calculated from the daily maximum 8-hour means.

was carried out with the EMEP model to calculate from the optimised emissions the ozone levels expressed as NET60[20], i.e. in the form of the target value.

B. Adaptation of model results to measured ozone levels

Since the RAINS model, like all models, is an imperfect representation of reality, deviations could be expected between the modelled ozone levels and the measured levels. These can only be quantified for past years for which measurement data exist. In order to take deviations between modelled and measured ozone levels into account as far as possible, a procedure was developed to adapt the modelled concentrations for the base years around 1990 to the measured concentrations in that period, and then apply this adaptation to the concentrations predicted for 2010 (Van den Hout and Roemer, 1999). This procedure allowed model/measurement mismatches in the base years to be corrected, so only the model uncertainty in the reduction of ozone levels between the base situation around 1990 and 2010 remained. The procedure is described in Annex C. Although the procedure attempts to correct model deviations, the results cannot be regarded as highly accurate. Two variants of the method, applied to explore the uncertainties, gave results that differed significantly, while the limited accuracy of some of the measurement data that could be used added to the uncertainties. The Steering Group discussed these limitations and recognised the uncertainties in the predictions of attainable targets in 2010. On the other hand, the majority of the Steering Group felt that this uncertainty essentially reflected the limitations of current scientific knowledge. Although improvements seemed possible, no significant reduction in overall uncertainty could be expected from continued, more extended investigation into this subject and so, to avoid delay, no attempt was made to refine the calculations. The consequences of the inherent imperfections of the models are likely to be most important for smaller countries. The Group recommends that this be born in mind during the formulation of the quantified obligations of the proposed ozone and national emission ceilings Directives. As for the ozone Directive a review mechanism should be included explicitly into the national emission ceilings Directive.

5.1.3. Optimised reductions of emissions and ozone

In the H1 scenario, chosen as the basis for the ozone reduction strategy, emission reductions and costs were derived from the following environmental objectives, which serve in the modelling as minimum criteria to be fulfilled in every grid cell:

1. Health-relevant ozone exposure:

    - Ceiling: AOT60 = 2.9 ppm.hours;

    - Gap-closure: reduce AOT60 everywhere[21] by at least two-thirds between 1990 and 2010.

2. Vegetation-relevant ozone exposure:

    - Ceiling: AOT40 = 13 ppm.hours[22];

[20] NET60 is defined as the number of exceedances above the threshold concentration of 60 ppb (120 µg/m^3) calculated from the daily maximum 8-hour means.

[21] I.e. in all grid cells of about 150 km x 150 km.

- Gap closure: reduce the excess AOT40 above the WHO guideline of 3 ppm.hours by at least one-third between 1990 and 2010.

3. Acidification:

    - Gap closure: reduce the area of unprotected ecosystems everywhere by at least 50% between 1990 and 2010.

4. Eutrophication:

    - No specific optimisation targets were defined for eutrophication, but the evaluation of environmental improvements associated with the scenarios took quantitative account of benefits in terms of eutrophication.

The H1 scenario yields a decrease of the exposure above the WHO guideline for human health and for vegetation (crops) by 76% and 53%, respectively, by 2010 compared to 1990 levels.

The two ozone ceiling levels are of direct relevance for the target values of the Ozone Daughter Directive, but the model targets could not be directly regarded as the real target values for 2010. After the optimisation calculations by the RAINS model, the two post-processing calculations mentioned in the preceding section had to be carried out for the health-related target value, whilst only the second calculation was needed for the vegetation-related target value. The results are described and discussed in Annex C. On the basis of the results the following numerical values are proposed for the target values:

- *Health-related target value: NET60 = 20-25 exceedance days on average over 3 years;*

- *Vegetation-related target value: AOT40 = 8-9 ppm.hours (16000-18000 $\mu g/m^3$.hours) on average over 5 years.*

These numerical values have been determined on the basis of the information available at the time of writing; substantial changes in e.g. the expected baseline situation defined in the REF scenario could be a reason to reconsider the values.

The optimised NO_x and VOC emission reductions per Member State in the H1 scenario are depicted in Figures 5.1 and 5.2 (Amann *et al*., 1999). The figures also show the reductions for the reference scenario REF, which represents the expected situation should no reduction measures be taken in addition to the current and planned measures. For comparison, the results for six other scenarios are indicated as well. The boxes mark the ranges spanned by a low and a high ambition scenario. The other symbols relate to four scenarios defined by way of sensitivity analysis (H4, H7, H8 and H12). For a more detailed explanation the reader is referred to Amann *et al*., 1999.

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[22] In the IIASA reports on this, the numerical value of AOT40 is expressed as the excess above the WHO guideline of 3 ppm. hours, so the ceiling level is expressed there as 10 ppm. hours.

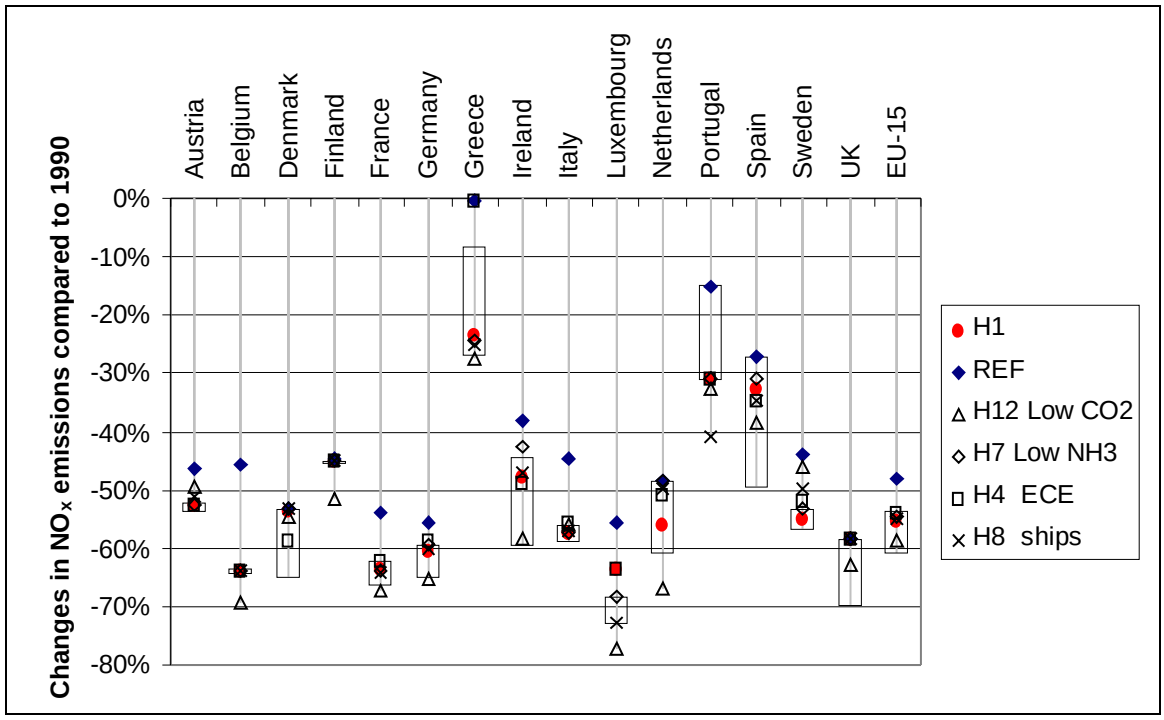

Figure 5.1 Comparison of NO_x emission reductions compared with 1990 (taken from Amann et al.*, 1999). The solid diamonds mark the percentage reductions of the REF scenario, the circles indicate the H1 scenario. The boxes show the range spanned by the low and high ambition scenario).*

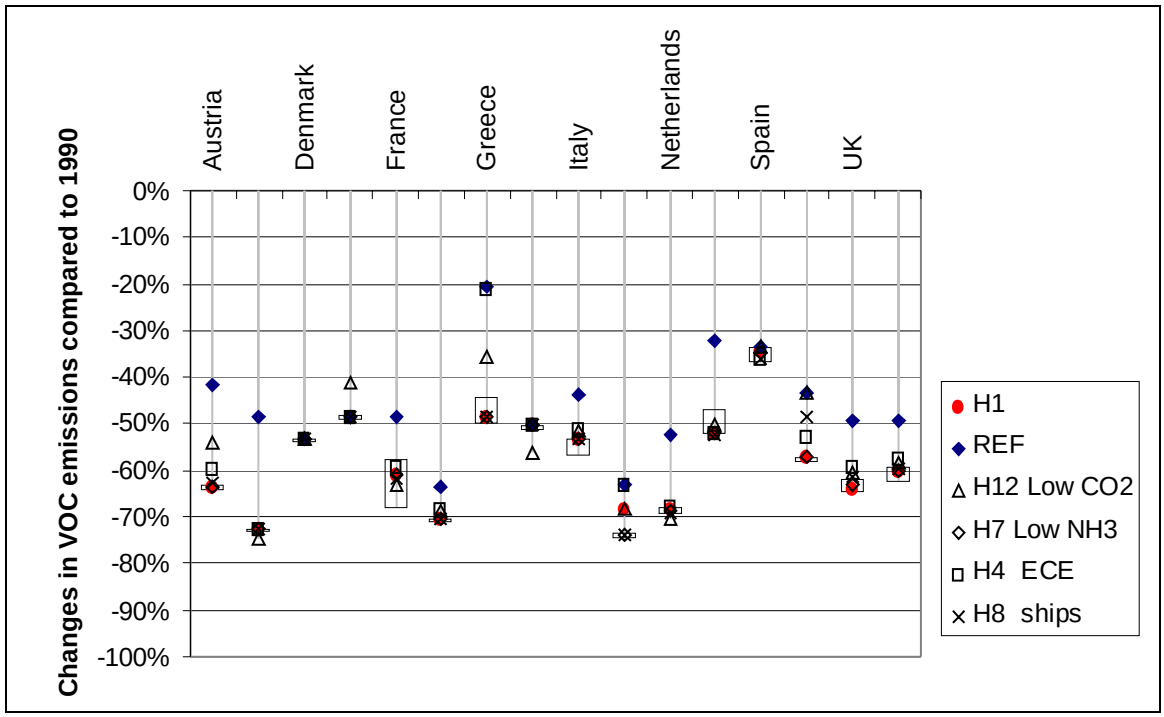

Figure 5.2 Comparison of VOC emission reductions compared with 1990 (taken from Amann et al., 1999). The solid diamonds mark the percentage reductions of the REF scenario, the circles indicate the H1 scenario. The boxes show the range spanned by the low and high ambition scenarios.

5.2. Cost-benefit analysis

5.2.1. Introduction

Chapter 3 proposes long-term ozone objectives based exclusively on assessment of the risks from ozone. Since it was recognised that these objectives could not be achieved in the foreseeable future, it was also proposed that target values be set that are expected to be attainable. The preceding sections of Chapter 5 summarise the development of these target values on the basis of available emission reduction technologies and associated costs. Parallel to this development a study was commissioned from AEA Technology to review the benefits associated with progress on air quality. This section summarises the results of this study (AEA Technology, 1998). The study attempted to express benefits in monetary terms to allow a comparison with the costs. Although the decisions on what environmental targets to set do not depend on the cost/benefit balance alone, the benefit estimates provide useful elements for use in consideration of the proposed target values.

5.2.2. Methodology

Benefits of the scenarios investigated using the RAINS model have, as far as possible, been calculated in monetary terms using the ALPHA model (AEA Technology, 1998) to allow them to be compared with estimated costs. ALPHA takes the RAINS emission scenarios and EMEP pollution modelling data on a 150 x 150 grid as inputs. For the cost-benefit study the 'ozone-only' H5 scenarios were used which have the same ozone targets as H1, H2 and H3 (see Section 5.1.3), but no targets for acidification. Consequently these scenarios only consider abatement of NO_x and VOCs and leave SO_2 and NH_3 untouched. Three variants were studied: H5/1 (low ambition), H5/2 (central) and H5/3 (high ambition) using results given in IIASA's 7th interim report. The EU emission reductions and costs for NO_x and VOCs in H5/2 were found to be almost equal to those in H1, indicating that the ozone-related targets, and not the acidification targets, are determining the emission reductions for these substances.

The benefits analysis then concentrated on effects on health, materials and crops. Effects on visibility and forest productivity were quantified but with very low confidence. It was not possible to quantify benefits to ecosystems beyond the assessment of exceedance of critical loads and levels already completed within RAINS. Analysis was not restricted to ozone effects: effects of the ozone precursor NO_x and its non-ozone secondary pollutants (particularly nitrate aerosols) were also considered. Effects of the other group of precursors, VOCs, were not considered because of insufficient detail in available emission inventories and the coarse scale of the analysis.

Health impacts dominate the overall results of the analysis though the study demonstrated that non-health effects were significant in the comparison with costs. So far as health was concerned the work covered: morbidity effects (such as respiratory hospital admissions and asthma attacks); effects on mortality from short-term exposure (often called acute mortality) to ozone pollution and nitrate aerosols; mortality effects from long-term exposure (often called chronic mortality) to nitrate aerosols. Chronic effects of ozone pollution were not considered due to lack of health impact data.

Given the inherent uncertainties of benefits assessments, a key part of this work was to assess the sensitivity of the results to alternative assumptions. The most important issue is probably the valuation of mortality impacts. For this, the study used two different approaches, value of a statistical life (VOSL) and value of a lost life year (VOLY), to indicate the potential range of results. Both techniques seek to assess individuals' (rather than "society's") willingness-to-pay (WTP) to reduce the risk of premature mortality. The VOLY approach assumes that life expectancy is relevant, whilst use of the VOSL does not. The result is an indicator of the importance that people attach to risk, and not an assessment of how valuable life is *per se*. Choosing a VOSL value for a particular study is difficult. The approach taken here used relatively conservative estimates of EUR 2.2 million as the VOSL for acute mortality effects and EUR 1.1

million for the VOSL for chronic mortality effects. There is concern that the VOSL will over-estimate WTP to avoid the risk of death linked to air pollution, because it is believed that the reduction in life expectancy attributable to exposure is small. This will often be the case for example, where pre-existing chronic respiratory or cardiac disease is a factor in death. The VOLY approach seeks to adjust for the short life expectancy of those affected. Here, values of EUR 110 000 per life year lost were used for acute mortality and EUR 67 000 for chronic mortality[23]. In the absence of consensus, the two approaches were used to indicate the sensitivity of the comparison of costs and benefits to this issue. Therefore, for both chronic and acute health effects we have two measures of the benefits. The lower estimates reflect the use of the VOLY approach. The higher estimate adopts the VOSL approach.

5.2.3. Costs

The Reference Scenario (REF) of the optimisation calculations by IIASA, based upon (the stricter of) current legislation and current reduction plans in Member States, provided the baseline against which all costs and benefits have been assessed. Table 5 presents a summary of the calculated costs of the REF scenario and the additional cost of attaining the combined targets of H1 and the ozone-only targets of H5/2.

Table 5.1 Emission Control Costs for the Joint Scenario H1 and the Ozone-Only Scenario H5/2 compared with the REF Case, in EUR million/year

	NO_x and VOC			NO_x, VOC, SO_2 and NH_3		
	REF	H1	H5/2	REF	H1	H5/2
Costs:	Total	on top of REF		Total	on top of REF	
Austria	902	119	120	1093	119	120
Belgium	1278	459	459	1704	1053	459
Denmark	484	0	0	623	6	0
Finland	642	0	0	889	0	0
France	7383	739	719	8659	916	719
Germany	10549	1048	933	13813	2147	933
Greece	1048	338	363	1482	338	363
Ireland	477	4	0	618	44	0
Italy	7868	403	420	9644	403	420
Luxembourg	71	4	30	98	4	30
Netherlands	1731	211	140	2267	971	140
Portugal	1349	57	57	1530	57	57
Spain	5658	13	10	6495	22	10
Sweden	1125	87	73	1554	87	73
UK	6695	1026	957	7964	1348	957
EU-15	47258	4508	4280	58433	7514	4280

Meeting the proposed target values for ozone is expected to require an additional reduction in VOC emissions of around 22%, and additional reductions in NO_x of around 12% relative to the Reference scenario. Large reductions in emissions from road transport are now included in the Reference scenario as a result of the conciliation process on legislation following the Auto-Oil I programme. So, the bulk of the cost of eliminating remaining problems is expected to derive from

[23] The figure for chronic effects is lower because chronic effects only arise many years in the future, and so are given a lower weight.

measures to be taken at (low stack) stationary sources. The cost of meeting the target values is estimated at EUR 4.3 billion per year in the EU. It is very likely that these cost estimates are on the high side because they have been based on "end of pipe" measures only, omitting cost-saving changes in energy supply or other structural changes which could be the preferred response of industry. For example, investigation of a "low CO_2" scenario suggested that the additional costs would fall by 40% if Community emissions of CO_2 came close to the Kyoto objectives.

On the other hand, no precise estimates can be given of the costs of any further efforts to combat locally induced ozone formation in order to comply with the target value (see Section 5.3). The close dependency upon very specific local circumstances (meteorology, local emission structure, photochemistry, etc.) makes it impossible to analyse which conurbations would require such additional local measures, let alone to estimate the costs of such actions. In any event, the sum of additional efforts on a local scale will remain small relative to the EU-wide strategy, and the overall costs of such measures will be a minor fraction of the total expenses.

5.2.4. Benefits

The estimated monetary benefits of the proposed targets in IIASA's H5/2 scenario range from EUR 3.3 billion to EUR 11 billion per year, if chronic effects on health are excluded. The large range here is mainly due to uncertainty in the valuation of acute mortality. The benefits of reductions in acute mortality range from EUR 180 million (using the VOLY) to EUR 7.3 billion (using the VOSL). The bulk of these benefits derive from reduced damage from fine particulate matter related to ozone precursors. Morbidity benefits account for EUR 1.2 billion, improved agricultural productivity for EUR 1.9 billion, materials benefits for EUR 17 million and forest benefits for EUR 140 million.

If chronic health effects are included in the benefit estimates, the total benefits are estimated to range from EUR 9.9 billion (using the VOLY) to EUR 18 billion (using the VOSL) per year. Again, the method used for valuation of mortality is the factor determining the wide range. The valuation of the other effects (morbidity, materials, agriculture, etc) remains the same.

5.2.5. Comparison of costs and benefits using sensitivity analysis

There is some debate about the robustness of the functions underpinning the assessment of chronic health effects. This is why the results are presented separately above. There are other uncertainties in the benefits analysis, at most levels of the modelling, but the most significant sensitivities overall are the approach to valuation and the inclusion of chronic effects. Other errors are unlikely to have as significant an effect on the balance between costs and benefits. For example, the inclusion of a threshold effect for acute mortality from ozone considerably reduces the direct benefits of reducing ozone concentrations. However, this has relatively little effect on the overall cost/benefit balance, as many of the health effects are related to the precursor gases and associated particulates.

In conclusion, benefits substantially outweigh costs under most combinations of assumptions, except where the VOLY approach is used to value mortality, and chronic effects are excluded. If chronic effects are included, then benefits clearly outweigh costs, irrespective of the way mortality is valued.

5.3. Strategies for the regional and urban scale

5.3.1. General

This section deals with the potential for local and regional measures to supplement large-scale measures. As described in Chapter 1 and the Consolidated Report (Beck *et al.*, 1999), it is known that ozone peaks can occur on the regional and urban scale, superimposed on the larger-scale background levels. Since these smaller-scale peaks are also subject to the provisions of the ozone

Directive, it should be investigated how they can be avoided, especially in urban situations, where many people are exposed.

The EU-wide strategy described in Section 5.1 focuses on the large-scale ozone levels. It is based on calculations of the EMEP/RAINS model with a resolution of 150 km, which do not resolve peaks of spatial dimensions below this size. Since the model assumes uniform measures per country, the potential of local measures in regions that are larger than 150 km but smaller than the Member State to which they belong, is also not addressed.

Furthermore, only durable emission reductions were taken into account in the EMEP/RAINS model calculations and so the possibilities of short-term measures have not been not dealt with, either for the local or for the large scale.

Large-scale measures affect the local situation in two ways:

1. EU-wide measures affect the background levels of ozone and of precursors.

   Since the EU strategy aims to reduce both ozone levels and precursor emissions, background precursor levels can also be expected to decrease. The ratios between precursors in background air flowing into cities will also change to some extent, although this systematic change will probably not be large compared with the day-to-day variations in the precursor mix. These changes are in principle addressed by the EMEP/RAINS model.

2. EU-wide measures change local emissions of precursors and thus the local contribution to ozone.

   The large-scale model calculations do not indicate how urban levels will change. Long-term average ozone levels *in* cities are almost always lower than *around* cities, but during episodes, when ozone is typically formed much closer to the sources than usual, city levels can be both lower and higher than background levels. These changes have been addressed in indicative calculations of the OFIS model (see below).

5.3.2. Local conditions and local ozone

In many NW-European cities, local episodic levels tend to be somewhat lower than in surrounding areas due to titration of ozone by NO_x emitted by local sources. In such situations, local NO_x reductions would not be expected to prove effective in reducing levels in the inner city. Local VOC emission reductions, on the other hand, can generally be expected to have an ozone-reducing effect in the city, even in the centre. In city plumes traversing the outskirts of cities and beyond, ozone levels are more likely to be increased compared with the background air. The balance between scavenging and formation depends on city size, meteorological conditions and the topographic structure of the area. In field studies both increases and decreases have been observed.

In Mediterranean regions the situation is even more complex. The (re)circulation of polluted air is very difficult to characterise due to the often very complex topography (mountains, often in combination with land-sea interface). Due to sea-breeze re-circulation, which is in many locations the norm rather than the exception, air can remain within a region for a prolonged period, and so formation due to local and regional sources is expected to be more prevalent around the Mediterranean Sea than in NW Europe.

Annex D gives an overview of current knowledge on the effectiveness of local emission reductions. Unfortunately the data are too scarce to develop a statistical picture of how local episodic ozone levels are above or below regional background levels and how they respond to urban emission changes. Model studies have given some insight for some cities, but even for Milan and Athens, the subject of several modelling studies, a quantitative picture in statistical terms did not emerge. The

complexity of current models makes it extremely difficult to develop statistics that are valid for cities in general.

To take a first step in this direction, the European Commission commissioned a modelling study (Moussiopoulos *et al.*, 1988) to develop statistics on ozone formation and its response to urban emission reduction for two example cities. In this study a simplified urban photochemical model, OFIS, was applied to Stuttgart and Athens. Emission reductions and air pollution levels resulting from the EU-wide scenario calculations of the EMEP/RAINS model (see Section 5.2) were taken as starting points for examining the effect of further local emission reductions, taken on top of the EU-wide emission reduction. The simplifications in the model made it possible to carry out simulations for each day over a six-month period (1 April to 30 September 1990). This extended period meant the results could be presented in statistical terms, including NET and AOT values. Lower ozone burdens were found in the case of one-sided VOC emission reductions. One-sided NO_x emission reductions on the other hand seemed to lead to more ozone in the urban area, while benefits could occur at fairly large downwind distances. The calculations suggested that additional local NO_x emission reductions of the order of 70-80% would be required for an acceptable reduction of the overall ozone burden. It was concluded that in Central European air sheds like Stuttgart it is very important to combine emission reduction measures at both the regional and the urban scale. To a lesser extent this conclusion was deemed to be valid for isolated areas like Athens, where local measures had a larger share in reducing the urban ozone load.

A further application of OFIS to 23 urban areas, carried out in the framework of other European studies, confirmed that the results for the city of Stuttgart might be transferred to the majority of EU urban areas.

Although this work added valuable information on the response of ozone to urban-scale measures in statistical terms, the techniques applied are not well enough developed to rely on the results in quantitative terms. It has to be concluded that the current state of the art does not allow us specify in general terms where and when local emission reductions are effective.

5.3.3. Possibilities for a generic approach

We can conclude from the above analysis that the effectiveness of specific local or regional measures depends greatly on local conditions. Current knowledge does not allow us to devise a generic set of local measures that will improve local ozone levels everywhere in Europe. This is true both for durable and short-term measures. Consequently, it does not seem feasible either for the ozone strategy to include a general strategy for specific local durable measures, or for the Ozone Directive to trigger specific local short-term measures that can be expected to be effective everywhere.

It is, however, desirable that throughout the EU in comparable situations the same obligation to consider local action should exist, and so we need to examine what possibilities exist to make provision for this in legal terms. Instead of prescribing specific measures at specific ozone levels, one could consider the option of prescribing local measures for cases in which such measures are sufficiently effective, without specifying these cases further. This would mean choosing an effectiveness criterion, which raises a new complication: the effectiveness of measures increases with the size of the area in which they are taken. Since for small areas emission reductions may even lead to local ozone increases, it is to be expected that the ratio between a measure's benefits and the efforts and costs it entails depends on the size of the area. Consequently, it does not seem feasible to fix a particular effectiveness criterion in terms of ozone reduction without taking the associated area into account.

There seem to be no practicable possibilities of defining a quantitative legal trigger for local actions. Since local actions are important elements in the abatement of ozone, it is instead

proposed that a procedure be defined which requires Member States to explore and assess the possibilities of local measures and to apply them if they can be identified as effective.

5.3.4. Types of local measure

Three levels of local engagement can be distinguished: (1) dissemination of information, (2) recommendations to the public and other sectors of society to reduce precursor emissions and (3) legally enforced reduction of emissions or of activities leading to emissions. (1) and (2) have been discussed in relation to the information and general alert thresholds in Chapter 3.

It is important to distinguish between durable measures, which are in force regardless of actual ozone levels and typically throughout the year, and short-term measures, which are only taken during or in anticipation of periods of high ozone.

5.3.5. Durable regional and urban measures

Annex E gives a list of durable regional and urban emission reduction measures that could be taken in addition to large-scale measures related to the EU strategy. Since they contribute to the achievement of national emission ceilings, they can be considered part of the measures within the EU strategy. Such steps should be reported to the Commission where the target value or long-term objective is exceeded, as proposed in Chapter 2. Consequently, special provisions for reporting regional and local measures are not needed.

5.3.6. Short-term regional and urban measures

Annex E lists short-term regional and urban emission reduction measures that could be taken in addition to the measures related to the EU strategy. Since ozone formation during episodic conditions takes effect closer to sources than during other conditions, the potential of short-term measures can be relatively high. Since it was judged not to be feasible to formulate short-term strategies at the EU level, an assessment is required of the effectiveness of such measures under local circumstances. The decision as to whether such actions should be taken could be the responsibility of national, regional or local authorities. Whether this decision process is mandatory depends on the local ozone levels (see Chapter 2). Where the general alert threshold is exceeded, the result of this process should be reported to the Commission. To avoid unnecessary work, a three-step procedure could be followed:

1. Investigate and report whether there is potential for regional or local short-term measures to reduce ozone. If a significant potential does exist, proceed to step 2.

2. Investigate, assess and report the effectiveness and feasibility of possible regional and local short-term measures, possibly including costs. If such measures are effective and feasible, proceed to step 3.

3. Develop an action protocol defining the conditions under which regional or local short-term measures are to be taken.

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6. REPORTING OZONE LEVELS

Member States should report the results of their assessments, in particular exceedances of thresholds. This chapter discusses aspects of various forms of reporting:

- Reporting to the Commission
- Reporting to the public
- Information exchange between neighbouring zones
- Reporting in relation to other frameworks
- Reporting in general

Information on causes and air quality management, to be reported in the event of exceedances of target values and long-term objectives, is not discussed here.

6.1. Reporting to the Commission

It is proposed to maintain the arrangements of the current ozone Directive 92/72/EEC for reporting to the Commission, both on a monthly basis and on an annual basis.

Monthly reports on maximum levels and threshold exceedances, to be sent before the end of the following calendar month, are needed in the summer half-year to provide a recent picture of the ozone situation in the EU, and to enable the Commission to issue a report on EU levels soon after the summer period. The option of reporting only once, at the end of August, on recent summer peak levels is not preferred, since it would not reduce the work needed for routine monthly reporting very much and would deny the Commission access to information on recent ozone levels. In some Member States monthly reports are already routinely produced for internal use. The use of automatic transmission of data can facilitate the monthly reporting. Requiring Member States to make the data available for downloading by the Commission was not considered a practicable option. It should be noted that the final data quality assurance may have to take place after the calendar year, so the data in the monthly report may have a preliminary status.

The annual reports containing statistical data are to be sent before 1 July of the following year. This differs from the deadline of 1 October for information to be sent to the Commission under the Exchange of Information Council Decision, but it was felt that in the case of ozone, with high levels only in the summer months, reporting in September would delay the Commission's report until the winter. (The data to be sent for the Exchange of Information Council Decision (time series of hourly data) are in fact different from those required for the Daughter Directive (statistical data)).

Table 6.1 lists the data to be submitted to the Commission in the framework of the Ozone Daughter Directive. As the monthly reports also include the maximum, it is necessary to report every month, irrespective of the levels observed. The precise form of the measured data to be reported is not specified here, having already been defined in the framework of the current Directive.

In Chapter 4 it is proposed that measurements of concentrations of NO_x and VOCs be carried out too. The purpose of these measurements is twofold: (1) NO_2 measurements collocated with ozone measurement are recommended to allow better characterisation of the representativeness of stations, and (2) measurements of NO_x and VOCs produce information from which precursor emissions and trends in these emissions can be derived.

Results from measurements for the first purpose should be reported in close connection and simultaneously with the ozone results.

The second purpose may lead to special measuring strategies not compatible with the assessment of air quality as such. The processing and reporting of these data may be very different than for data from regular air quality monitoring stations - in fact, it may cause confusion to report data with anomalous representativeness. Currently it does not seem feasible to detail the way in which precursor measurements should be reported. It is recommended that a common framework be developed during the implementation of the ozone daughter directive.

The reports on observed levels at monitoring sites, which are essentially point measurements in space, are far from providing a complete, territory-covering overview of ozone levels in the zones of the Member States. In the "Consolidated Report" (Beck *et al*., 1999) attempts have been described to extrapolate monitoring results to the entire EU territory. Since the representativeness of the monitoring sites was not well known, it was found to be impossible to construct a reliable map of ozone over Europe. Also, after implementation of the Daughter Directive it will be difficult to generate ozone maps centrally on the basis of monitoring results alone, because of the difficulties of taking special local conditions into account, e.g. the ozone gradients in mountainous areas in Austria. Because these are best known and interpreted locally, Member States should be encouraged to report not only ozone levels at stations, but also maps of ozone derived from the measurements using mathematical techniques such as extrapolation and modelling. It would be very useful to develop a common standardised form for reporting such results for the future Daughter Directives. A reporting format for concentrations should be developed that includes, besides statistics on temporal concentration distribution, information on spatial concentration distribution in the zones. This may include maps and statistical parameters on the spatial concentration, analogous to the temporal statistics now being reported by monitoring stations. Information on population exposure may also be included.

In addition to this, information on other assessments could be collected at the European level, in particular by the ETC-AQ: modelling results, trend information, evaluation of episodes, etc. However, it does not seem feasible to develop a standardised format for this type of information.

6.2. Reporting to the public in the event of exceedance of the Information or General Alert Thresholds

In Chapter 3 it is proposed that information be released to the public when an information or general alert threshold is exceeded or expected to be exceeded. Annex F gives proposals for harmonising the messages in the form of a list of items that should be addressed as a minimum in the information text. While there are arguments in favour of standardising the texts to be issued to the public, mandatory texts are not preferred because of contextual differences that may exist between Member States.

In addition, Member States should make general information on the ozone problem available to the public, as background and reference material.

6.3. Exchange of Iiformation between neighbouring zones in the event of exceedance of the Information or General Alert Threshold

It is recommended that a regular system be established for informing neighbouring zones of observed or forecast exceedances of the information or general alert thresholds.

6.4. Reporting in relation to other frameworks

The Commission intends to propose the daughter directive on ozone together with the National Emission Ceilings (NEC) Directive, which constitutes the keystone of the ozone abatement strategy. As discussed in Chapter 5, there is a close relationship between the national emission ceilings and the ozone target value to be proposed. In view of the considerable effort and high cost involved in reaching the targets proposed for both ozone and the national emission ceilings, it is of

great importance to the Commission, the Member States and other stakeholders to follow the progress of the implementation of the abatement strategy, and its effects on ozone concentrations.

In the framework of the Convention on Long-Range Transboundary Air Pollution, as in the Commission, there is growing interest in monitoring implementation of abatement strategies. In 1997, an Implementation Committee was established that will:

- periodically review compliance with reporting requirements,
- ensure that the quality of information submitted has been evaluated
- consider problems arising during implementation and suggest constructive solutions
- prepare a report on implementation

EMEP is now considering including an explicit implementation aspect in its monitoring strategy, by setting up monitoring aimed at verification of emissions and emission trends. This concurs with the proposals in the EC ozone framework to include monitoring of ozone precursors for the same purpose. This could provide independent evidence on the validity of emissions as obtained from emission inventories. A recent UK study has demonstrated the feasibility of this approach for VOCs (Derwent *et al*., 1998*)*, while a Dutch study has evaluated trends in measured NO_x concentrations in relation to trends in concentrations calculated on the basis of emissions (de Paus *et al*., 1997).

It is proposed that a regular report be produced, combining and interrelating information on:

- ozone exceedance of target levels;
- progress in ozone abatement;
- attainment of precursor emission targets.

Aspects to be covered in the report could be:

- recent developments in economic sectors that are important for ozone precursor emissions;
- updated precursor emission estimates and emission trends as obtained from inventories
- verification of these emissions and trends from precursor measurements in ambient air;
- measured ambient ozone concentrations and exceedances;
- updated information on population exposure;
- checking whether observed ozone concentrations and exceedances are as expected given the emission trends (evaluation with RAINS model);
- Member States' progress in implementing measures;
- information on the actual costs of measures (in relation to earlier cost estimates)

Considering the reporting requirements under the air quality framework Directive, a reasonable frequency of reporting these aspects could be three years. This reporting could be carried out within the triennial report provided for in Article 11(2)(b) of the Framework Directive.

6.5. Reporting in general

Apart from reporting to the Commission and to the public as discussed on Sections 6.1 and 6.2, Member States should also make reports on monitoring results available to the public, the media and the scientific community. The way in which this is done is not to be specified in detail, but for reasons of harmonisation it is recommended that a minimum set of data be defined, for publication. In this way information on ozone levels can easily be exchanged between Member States, and comparisons are facilitated. In earlier directives a minimum threshold was proposed for the obligation to issue this type of information, but in the case of ozone it is better to make this information always available. It is important to have a complete picture of ozone levels across the entire EU because ozone is a widespread problem with important long-range and transboundary contributions.

Obviously, exceedances of the target values, long-term objectives and information and general alert thresholds should be part of the data given in annual reports, daily information on the internet, teletext etc.; legal provisions on dissemination have been described above. Table 6.2 adds to this three other important reference levels to be mentioned in reports in order to facilitate international comparison and help the reader interpret ozone levels: threshold levels for visible damage to vegetation, damage to forests and damage to materials (see also discussions of these thresholds in Chapter 3). The last column gives a recommended frequency of publication. Publication on a daily basis refers to electronic media such as Internet and teletext, and publication with lower frequency includes annual reports.

Table 6.1 Data on ozone and ozone plus NO_2[1] to be submitted to the Commission (Supplementary to the exchange of information Council Decision)

Objective	Type of station	Pollutant	Parameters to report every month from April to September	Parameters to report for each year
Information threshold	Any	Ozone; ozone plus NO_2	- For each exceedance day: date, hour(s) of exceedance; for hour of maximum ozone: ozone and ozone plus NO_2 concentration; - Monthly 1h max of ozone	For each exceedance day: date, hour(s) of exceedance; for hour of maximum ozone: ozone and ozone plus NO_2 concentration[6]
General alert threshold	Any	Ozone; ozone plus NO_2	Same as above	Same as above
Health protection indicator	Any	Ozone; ozone plus NO_2	For each exceedance day: date; for 8h period of maximum ozone: ozone and ozone plus NO_2 concentration	For each exceedance day: date; for 8h period of maximum ozone: ozone and ozone plus NO_2 concentration
Vegetation protection indicator	Suburban rural, rural background	Ozone	Once in September: $AOT40_{veg}$	$AOT40_{veg}$[2]
Indicator for protection against visible injury	Suburban rural, rural background	Ozone	-	Max, 98^{th}, 50^{th} percentile of running $AOT40_{visdam}$[3] values
Indicator for forest protection	Suburban rural, rural background	Ozone	-	$AOT40_{forest}$[4]
Indicator of materials protection	Any	Ozone	-	Annual mean
General information	Any	Ozone; Ozone plus NO_2	-	maximum, 99.9^{th}, 98^{th}, 50^{th} percentile and number of valid data constituting the hourly series
General information	Any	Ozone; ozone plus NO_2	-	maximum, 98^{th}, and 50^{th} percentile from series of daily 8-hour maxima and number of valid data constituting that series.
General information		Nitrogen dioxide (NO_2)	-	Annual mean and number of valid data constituting hourly series
General information		Nitrogen oxide[5](NO_x)	-	Annual mean and number of valid data constituting hourly series

[1]Ozone plus NO_2 added as parts per billion and expressed in µg/m³ ozone equivalents (only for stations with ozone and NO_2 measurements).

[2] The indicator for vegetation protection $AOT40_{veg}$ is defined as the sum of the differences between the hourly ozone concentrations in µg/m^3 and 80 µg/m^3 for each hour when the concentration exceeds 80 µg/m^3 during the period 8.00h-20.00h in the months May-July.

[3] The indicator for protection against visible damage $AOT40_{visdam}$ is defined as the sum of the differences between the hourly ozone concentrations in $\mu g/m^3$ and 80 $\mu g/m^3$ for each hour when the concentration exceeds 80 $\mu g/m^3$ during five consecutive calendar days. The value is assigned to the last of the five calendar days.

[4] The indicator for forest protection $AOT40_{forest}$ is defined as the sum of the differences between the hourly ozone concentrations $\mu g/m^3$ and 80 $\mu g/m^3$ for each hour when the concentration exceeds 80 $\mu g/m^3$ during the period 8.00h-20.00h in the months April-September.

[5] Nitric oxide and nitrogen dioxide added as parts per billion and expressed in $\mu g/m^3$ nitrogen dioxide equivalents (only for stations with ozone and NO_x measurements).

[6] Exchange already required by Council Decision 97/101/EC.

Table 6.2 Reference Levels for Ozone related to Effects concerning Materials and Visible Damage to Crops and Vegetation

Target	**Reference level**	**Type of station**	**Recommended frequency of publication**
Visible damage to crops and vegetation	$AOT40_{visdam}$ = 400/1000 $\mu g/m^3$ h	Station targeted at protection of vegetation	Monthly, yearly
Damage to forests	$AOT40_{forest}$ = 20 000 $\mu g/m^3$ h	Station targeted at protection of vegetation	Yearly
Damage to materials	Annual average = 40 $\mu g/m^3$	Any	Yearly

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R. G. Derwent, T. J. Davies, M. Delaney, G. J. Dollard, R. A. Field, P. Dumitrean, P. D. Nason, B. M. R. Jones and S A. Pepler (1999). Analysis and interpretation of the continuous hourly monitoring data for 26 C2-C8 hydrocarbons at twelve United Kingdom sites during 1996. Atmospheric Environment, in press.

de Paus, T.A., Roemer, M.G.M., (1997), Trends of NOx in North-West Europe, Report TNO-MEP-R97/136, TNO, Apeldoorn, the Netherlands.

ANNEX A: STRUCTURE AND MEMBERS OF THE AD-HOC WORKING GROUP

For the purpose of drafting this Position Paper on Ozone and supporting the Commission in its task of devising an ozone strategy and drafting a proposal for a daughter Directive on ozone, the "Ad-Hoc Working Group on Ozone Directive and Reduction Strategy Development" was set up in February 1997. For the sake of efficiency, the "Plenary" group decided to form two subgroups: the "Subgroup on Risk Assessment", which was concerned mainly with drafting chapter 3, and the "Subgroup on Monitoring Strategies", which drafted chapter 4. The Plenary Group and the Subgroup on Risk Assessment were chaired by the Commission, DGXI.D.3, while the subgroup on Monitoring Strategies was chaired by the Joint Research Centre, Ispra. The following table lists the members of the Ad-Hoc Working Group ('P' = Plenary Group) and their involvement in the work of the subgroups ('M' = Monitoring Strategies, 'R' = Risk Assessment).

Name	Institution	Group
Mr. Martin Lutz	European Commission, DGXI.D.3 Chairman of the Plenary Group and of the Subgroup on Risk Assessment	P,R,M
Mr. Emile De Saeger	Joint Research Centre (JRC) ,Ispra, Italy Chairman of the Subgroup on Monitoring Strategy	P,M
Mr. Roel van Aalst	European Topic Centre on Air Quality, RIVM (NL)	P
Mr. Christer Ågren	Acid Rain Secretariat (Sweden) (for EEB)	P
Mr. Markus Amann	International Inst. for Applied Systems Analysis (Austria)	P
Ms. Sylvia Baldinger	Ministry of the Environment (Austria)	P
Mr. Nelson Barros	University Aveiro, Dept. of Environment and Planning (Portugal)	M
Ms. Ruth Baumann	Umweltbundesamt (Austria)	P,M
Ms. Suzie Baverstock	CONCAWE (for UNICE)	P,R
Ms. Jeanette Beck	European Topic Centre on Air Quality, RIVM (NL)	P,M,R
Mr. Ton Blom	Ministry of Environment (The Netherlands)	P,R
Ms. Filomena Boavida	Instituto de Meteorologia (Portugal)	P
Ms. Annette Borowiak	JRC (Ispra)	P,M
Mr. Peter. Borrell	EUROTRAC	P
Mr. Brian Brangan	Department of the Environment (Ireland)	P
Mr. Keith Bull	Institute of Terrestrial Ecology (UK); Chairman of the UN-ECE Working Group on Effects	P,R
Mr. Gerwin Dumont	IRCEL (Belgium)	P,M
Mr. Bert Elshout	Kema NL. B.V (for UNICE)	P
Mr. Ola Glesne	Norwegian Pollution Control Authority	P
Ms. Annabelle Guivarc'h	Ministry of the Environment (France)	P
Ms. Anette Hauer	European Environment Bureau (EEB)	P,R
Mr. Mike Hawkins	Ford / ACEA (for UNICE)	P,M
Mr. Alain Henriet	PSA Peugeot Citroen/ ACEA (for UNICE)	P,M
Mr. Mike Holland	AEA Technology (UK), Consultant to the Europ. Comm.	P
Mr. Dick van den Hout	T.N.O. (NL), Consultant to the European Commission	P,R, M
Mr. Dieter Jost	Umweltbundesamt (Germany)	P
Mr. Duncan King	CONCAWE (for UNICE)	R
Mr. Harald Kohl	Federal Ministry of the Environment (Germany)	P
Mr. Antti Kulmala	Finish Meteorological Institute	M
Mr. Tarja Lahtinen	Ministry of the Environment (Finland)	P
Mr. Duncan Laxen	Air Quality Consultants (U.K. for EEB)	P,R
Mr. Rolaf van Leeuwen	World Health Organisation, Europe	P,R

Mr. Lars Lindau	Swedish Environment Protection Agency, Chairman of the UN-ECE Working Group on Abatement Techniques	P
Mr. Millán Millán	Centro de Estudios Ambientales del Mediterráneo (Spain)	P,M
Mr. Luis Duran Montejano	Iberski (Spain)	M
Mr. Finn Palmgren Jensen	National Environment Research Institute (Denmark)	M
Mr. John Pearson	Shell/CEFIC (for UNICE)	P,M
Mr. Håkan Pleijel	Swedish Environment Institute (for EEB)	R
Mr. Bernhard Prinz	Landesumweltamt Nordrhein-Westfalen (Germany)	P
Mr. Werner Rudolf	Umweltbundesamt (Germany)	M
Mr. Peter Rombout	National Institute for Public Health and Environment (RIVM, NL)	P,R
Mr. Franco de Santis	CNR – Istituto Inquinamento Atmosferico (Italy)	P,M
Ms. Maria Jose Sanz	Centro de Estudios Ambientales del Mediterráneo (Spain)	P,R
Mr. Nigel Sarginson	Chairman of UNICE Ozone Shadow Group (for UNICE)	P
Mr. Jürgen Schneider	Umweltbundesamt (Austria)	P,M
Mr. David Simpson	Norwegian Met. Institute, MSC-W (NOR)	P
Mr. Andreas Skouloudis	JRC (Ispra)	M
Mr. Remy Stroebel	A.D.E.M.E. (France)	P,M
Mr. Per Suhr	Danish EPA	P
Mr. Frank Thewes	Administration de l'Environnement (Luxembourg)	P
Ms. C.-M. Tomazinaki	Ministry of the Environment (Greece)	P, R
Ms. Maria Tombrou	European Topic Centre on Air Quality, Univ. of Athens, Dept. of Applied Physics	P,R
Mr. Martin Williams	Department of the Environment (UK), Chairman of the EMEP Steering Body	P,R
Mr. Henning Wuester	UN-ECE Secretariat	P
Mr. Ioannis Ziomas	University of Thessaloniki, Laboratory of Atmosph. Physics	R
Mr. Andre Zuber	Swedish Environment Protection Agency	P,R

The following group members provided larger text contributions or data evaluation results: Roel van Aalst, Markus Amann, Suzie Baverstock, Jeanette Beck, Annette Borowiak, Keith Bull, Gerwin Dumont, Luis Duran Montejano, Mike Holland, Dick van den Hout, Dieter Jost, Rolaf van Leeuwen, Martin Lutz, Millán Millán, Håkan Pleijel, Peter Rombout, Emile de Saeger, Maria Sanz and Andreas Skouloudis. Mr. Kucera (Swedish Corrosion Institute) provided the paragraphs on damages to materials. Michel Houssiau (consultant to the Commission) performed additional data analysis.

ANNEX B: OZONE IN MOUNTAINOUS REGIONS AND IN SOUTHERN EUROPE

The complex interplay between chemical and meteorological processes makes ozone formation difficult to understand. This is especially true for regions of complex topography. In coastal regions around the Mediterranean Basin, for instance, the combination of mountains and sea breeze re-circulations significantly affects ozone phenomenology. Ozone can also have very specific distributions in mountain areas, and observed concentrations differ significantly between mountain peaks and valleys. This annex gives a short overview.

Southern Europe[24]

In 1973 the European Commission began supporting research into the physico-chemical processes which govern the dynamics of air pollutants in various regions of Europe. In particular, six European Remote Sensing Campaigns were instrumental in documenting that polluted air masses were mostly advected in North-Western Europe, i.e. Belgium (Ghent), France (Cordemais) and in the UK (Drax), while they showed marked diurnal oscillation cycles in the airsheds of Southern Europe, i.e. France (Lacq, Fos-Berre) and Italy (Turbigo). The hypothesis that pollutants might be re-circulated in some regions of Southern Europe formed the basis of other EC projects dealing with the dynamics of photo-oxidants in the Mediterranean Basin. In these projects, air pollutants were regarded as tracers of opportunity of atmospheric flows in mid-summer (July).

Thus more knowledge is now available about specific meteorological processes in the Mediterranean and their links with one another from the local to the sub-continental scales. Other EC-supported projects have further documented that ozone formation in summer varies greatly across Europe, and even within the Mediterranean Basin. For example, the Western Mediterranean Basin is surrounded by high mountains and, in summer, is under weak levels of anticyclonic subsidence and strong insolation. These conditions favour the development of meso-scale processes and the re-circulation of air masses. During the same period the Eastern Mediterranean Basin is under conditions of weak ascent and strong advection, i.e. the Etesian winds, and the development of re-circulations is largely inhibited.

The Western Mediterranean Basin is surrounded by mountains of 1 500 m or more. On summer days their east- and south-facing slopes are strongly heated and act like orographic chimneys, favouring the early formation of up-slope winds that reinforce the sea breezes and link the surface winds directly with their return flows aloft, and further, with their compensatory subsidence over the sea. The result is the formation of stacked layers along the coasts with the most recently formed layers at the top and the older ones near the sea. These reach 2 to 3 km in depth, have variable width over land (up to 100 km), and extend more than 300 km over the sea.

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[24] A list of references is given in Annex 3 of:

J.P. Beck, M. Kryzanowski and B. Koffi (1998), Ozone in the European Union. "The Consolidated Report" by European Topic Centre Air Quality, Bilthoven, The Netherlands. European Commission, Office for Official Publication, ISBN 92-828-5672-0.

on which this section is based. The figures have been taken from:

Millan M.M., E. Mantilla, R. Salvador, A. Carratala, M.J. Sanz, L. Alonso, G. Gangoiti and M. Nazavo 1999: Ozone cycles in the Western Mediterranean basin: Interpretation of monitoring data in complex coastal terrain, J. Appl. Meteo. (In press).

The layers act as reservoirs of aged pollutants, and the lower ones can be brought inland by the sea breeze of the next day(s), creating re-circulations, as illustrated in Figure 1. Tracer experiments on the Spanish East coast have shown that turnover times range from 2 to 3 days. During the night the land-based processes die out, and the reservoir layers can drift along the coasts and contribute to regional, inter-regional and long range transport of aged pollutants. Similar processes involving re-circulations and/or oscillations of the aged air masses have also been documented in the Central Mediterranean. Important differences are known to exist between the Western and Eastern Mediterranean Basin.

At the larger scales, deep convection in some regions (e.g. Spanish and Turkish central plateaux), or strong up-slope winds in others (e.g. Alps and Atlas mountains), can inject aged air masses directly into the mid-troposphere (3 to 6 km) and into the upper troposphere (10(+) km), where they can participate in long-range transport processes within Southern and Central Europe, and at the continental-global scales, respectively.

Under strong summer insolation, the coastal re-circulations become "large natural photo-chemical reactors" where most of the NO_x emissions and other precursors are transformed into oxidants, acidic compounds, aerosols and O_3, leading to exceedance of EC thresholds (Figure 2). Relevant aspects of this problem are: (1) that the concept of ‘upwind’ (background) and ‘downwind’ (polluted) of conurbations is inappropriate in regions of complex coastal terrain where re-circulation processes are important, (2) that ozone is generated at the regional scale from emissions in urban centres and other NO_x source areas, and (3) that as much as 60%, or more, of the observed O_3 at any one site may result from advection within the recirculating air masses.

These situations are the norm, rather than the exception, for the coastal regions surrounding the Western Mediterranean Basin, and illustrate the existence of chronic-type O_3 episodes, created by atmospheric re-circulations, as compared with the peak-type episodes in Central and Northern Europe, which are created by combinations of long-range transport and atmospheric stagnation. They also reveal a problem of data interpretation for those responsible for monitoring networks from the local to the EC level.

Thus, in Southern Europe and the Mediterranean Basin the observed O_3 cycles depend strongly on the topographic location of the observing station and its relationship to the reservoir layers, the atmospheric circulations involved, and the chemical processes along each path, as illustrated in Figures 1 and 2. As a result, each O_3 monitoring station shows a part of the whole, and could even be considered to represent a specific area, providing the relevant processes are understood for each site and the site itself has been adequately selected; however, no single station can be considered representative of regional processes, and much less of the whole situation.

Mountain regions

High mountain sites

At altitude, concentrations can be high and fairly constant due, among other factors, to enhanced exchange processes with (the upper) reservoir layers which advect O_3 from source regions upwind. In turn, these layers may have been formed by orographic and/or deep convective injection, i.e. by natural venting of the surface layer. It should be emphasised that convective and/or orographic injection can be quite deep over some regions of Europe. Documented cases include, for example, from 3.5 to more than 5 km over the Spanish central plateau, and more than 5 km over the Alps. This injection is followed by stratified transport during the night, which means these layers can affect high altitude sites located far downwind.

In response to these mechanisms, the observed O_3 at high altitude sites can show a nearly flat cycle with high average values day and night. It may also show a minimum in the morning, at about the same time that a strong rise in O_3 is observed at stations located on the valley floor. Both of these are indicative of the development of convective mixing processes along the sun-heated valley walls, which include two complementary mechanisms, viz. the mixing of O_3-depleted air from the valley bottom with the layers

aloft, and the fumigation to the valley floor of O_3-enriched air from reservoir layers trapped within the valley during the night.

Valley sites

Processes affecting mountain peaks are also linked to the O_3 cycles observed in the valleys below. These tend to be dominated by the local wind regime under weak synoptic conditions, or by aerodynamic rotors and other orographic interactions with the upper winds (i.e. channelling) when the synoptic conditions are stronger, or by varying combinations of these.

Under weak synoptic conditions the observed O3 cycles can be quite complex in response to the following processes:

(1) During the night a stable surface layer forms by radiational cooling and (mostly) by the accumulation of cold drainage winds from the valley slopes. This airmass may be more or less shallow depending on whether it stagnates or whether a generalised down-valley drainage flow develops and becomes progressively depleted in O_3, particularly down-valley from any conurbations. Thus, down-valley stations may show very low O_3 values during the night due to deposition and/or titration by NO. Stations located on the valley floor and up-valley may show varying residual levels of O_3 from the drainage flows which still bring some O_3 from the (middle and upper) reservoir layers aloft. Finally, stations at any height along the valley walls will show varying O_3 values, depending on whether they remain inside the middle reservoir layers or are engulfed by the surface layer.

(2) During the morning a reversal of the drainage flow takes place. Fumigation of O_3 from the middle reservoir layers begins to occur just before or about the time of the reversal, and the observed cycles may include a sharp O_3 rise at all stations in the valley due to fumigation from the middle and upper reservoir layers. Following this, the observed cycles vary as a function of the station's location *vis-à-vis* the conurbations. The fumigation rise can be followed by a decrease of O_3 just up-valley (and by now downwind) from any conurbation. In other locations further up-valley, the fumigation can be followed directly, or with some delay, by a second rise in O_3 from photochemical production within the valley. Stations down-valley may show the initial rise due to fumigation and, if the mixing is maintained, a nearly constant value of O_3 until the evening, when flow reversal occurs again along with the formation of a new stable surface layer.

(3) Photochemical production of new O_3 within the valley and its transport both along the valley floor and up the heated valley slopes. The latter can produce new reservoir layers aloft, some of them directly over the valley floor (trapped layers), and others over the valley ridges (upper layers) whenever the slopes are sufficiently heated. By late afternoon these processes decay and the cycle starts anew.

Large cities located in valleys can strongly perturb the local wind regime as a result of: increased roughness, heat island effects, or both. The results can be: (a) blockage of the drainage flow up-valley from the city during the night, which becomes more stagnant than if the city was not there, (b) weakening or stagnation of the flow down-valley from the city, and even including the possibility of a flow towards the city just down-valley from it. During the day, the presence of the city could reinforce the up-valley wind component down-valley from the city and weaken it up-valley from the city. These effects vary from valley to valley and could significantly alter the general picture discussed above.

Figure 1. (see next page). Sketch of diurnal circulations in the coastal regions of the Mediterranean Basin in summer. The entrance of the sea breeze during the day and the formation of stratified (reservoir) layers aloft is illustrated, with letters (a) to (d) indicating successive stages in this process. The lower part (DAY) shows a schematic of the ozone decay, and subsequent production, as it interacts with NO emissions in a coastal city (Derwent and Davies, 1994). The nocturnal conditions illustrate the formation of drainage flows and the accumulation of a stable airmass at the bottom of the valleys and over the coastal plains. The draining airmass can become blocked at some distance from the coast whenever the sea surface temperature is higher than that of the air. The lower part (NIGHT) shows a schematic of ozone evolution along the path of the draining air. In these processes, stations located high above the coastal plains (#5) can remain within the reservoir layers during the night. The number codes correspond to the stations in Figure 2.

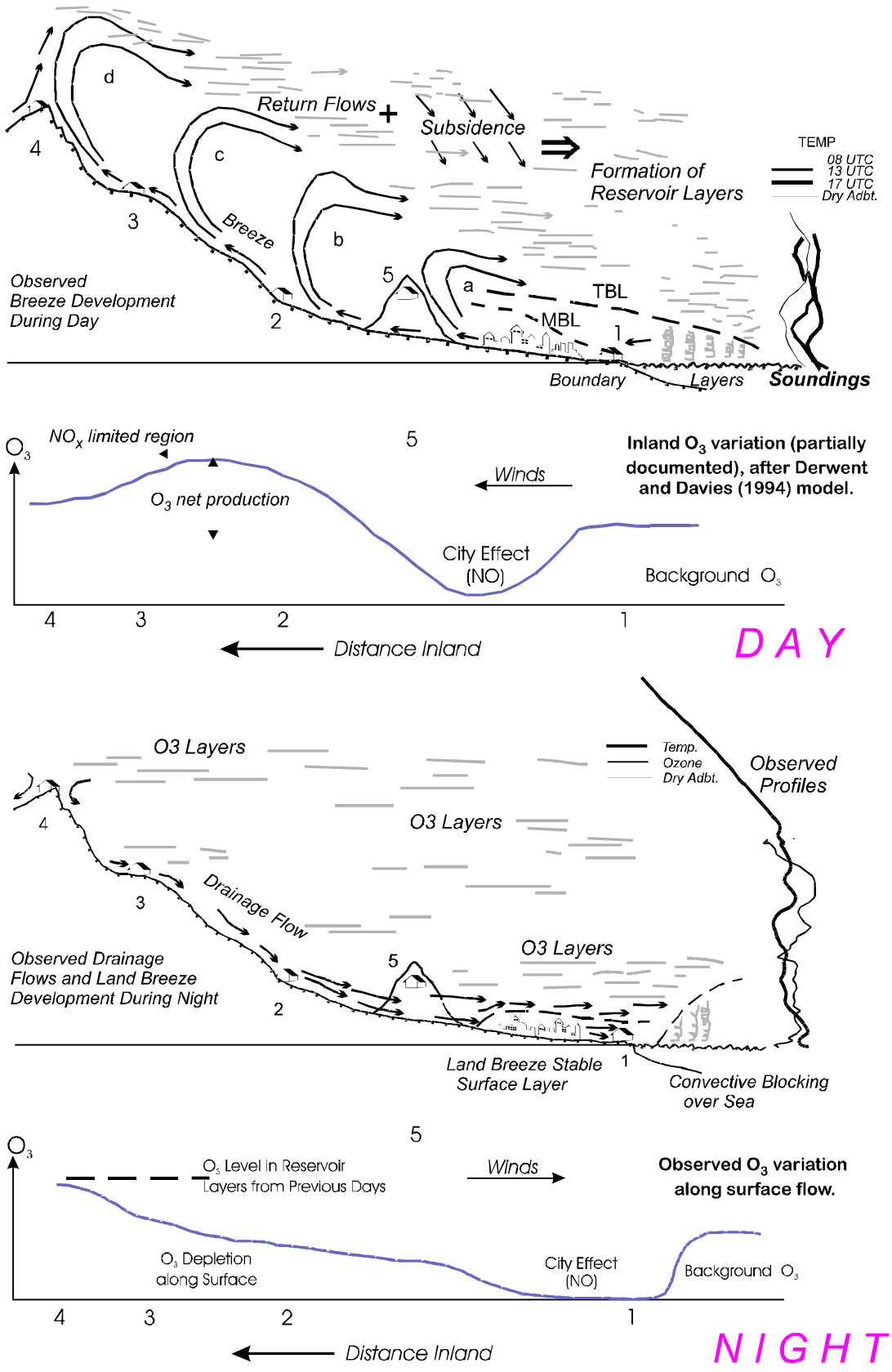
d
Return Flows
+
Subsidence
⇒
Formation of
Reservoir Layers
TEMP
08 UTC
13 UTC
17 UTC
Dry Adbt.
4
c
3
Breeze
b
5
a
TBL
MBL
1
2
Observed
Breeze Development
During Day
Boundary
Layers
Soundings
O3
NOx limited region
5
Winds
Inland O3 variation (partially documented), after Derwent and Davies (1994) model.
O3 net production
City Effect
(NO)
Background O3
4
3
2
1
Distance Inland
DAY
O3 Layers
Temp.
Ozone
Dry Adbt.
Observed
Profiles
4
O3 Layers
3
Drainage Flow
Observed Drainage
Flows and Land Breeze
Development During Night
5
O3 Layers
2
Land Breeze Stable
Surface Layer
1
Convective Blocking
over Sea
O3
5
O3 Level in Reservoir
Layers from Previous Days
Winds
Observed O3 variation
along surface flow.
O3 Depletion
along Surface
City Effect
(NO)
Background O3
4
3
2
1
Distance Inland
NIGHT

Figure 1

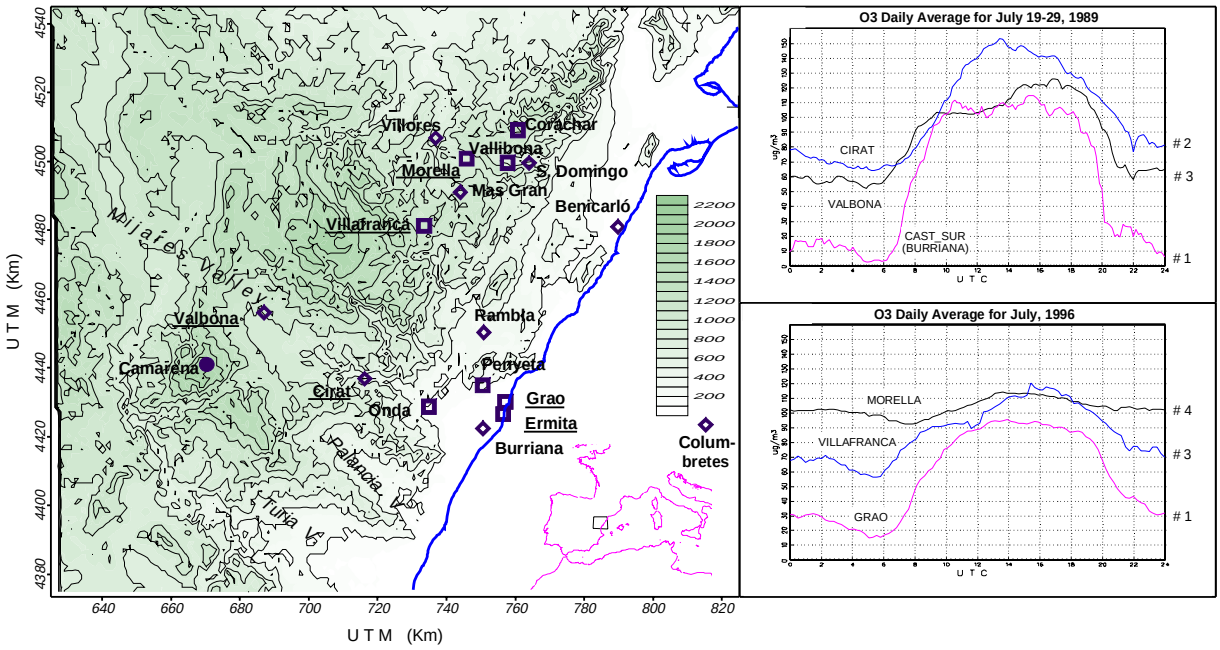

Figure 2. Daily ozone cycles in July for six stations located at various heights and distances inland from the Spanish East Coast. At the coast (Grao, Burriana, type #1) the O_3 drops to low values during the night and rises sharply between 0700 and 1000, just before the onset of the sea breeze. This is associated with the fumigation of O_3 from the reservoir layers. After this time it remains nearly constant and can be regarded as the background O_3 entering with the sea breeze. Cirat (type #2) is within the ozone production region. In Villafranca and Valbona (type #3), some O_3 is available from the reservoir layers, and concentrations do not drop below 60 $\mu g/m^3$ during the night. After the morning rise the O_3 remains nearly constant until the arrival of the sea-breeze front at 1200 and 1400, which produces a second rise in O_3 and the diurnal maximum by 1600. Finally, the mountain site (Morella, type #4) remains within the reservoir layers during the night. The O_3 remains high, its daily cycle is dampened, and it shows a minimum by 0800 which, not surprisingly, coincides with the time of maximum rise at the sites on the coast and valley floor.

ANNEX C: RELATING EMEP/RAINS SCENARIO RESULTS TO MEASURED OZONE LEVELS

Introduction

The target values in the forthcoming Daughter Directive on ozone will be based on model calculations by the EMEP and RAINS model. Since models are imperfect approximations of reality, it is important to take discrepancies between the model approximation and reality into account. This Annex describes calculations to adapt EMEP/RAINS model results for 2010 to measurements (Van den Hout and Roemer, 1999). While the calculations link the model results closer to measured ozone levels, a number of uncertainties remain. Two variants of a method are applied, one of which reproduces the model results reasonably well, whilst the other gives systematically lower reductions of ozone. On the basis of the results a proposal is given for the numerical value of the ozone target values.

Methodology

General approach

The model scenario results for 2010 will be the basis for the target values, but these results can obviously not be directly compared with measurements - only measurements of the past exist. Since model results for past years were only available for the period around 1990, the method will use measurements from this period.

In the approach chosen to select the target value:

- *measurements are taken to determine ozone levels in 1990;*
- *model predictions are taken to determine the reduction of ozone levels between 1990 and 2010.*

Measurements:	***Level in 1990***
Model:	***Reduction between 1990 and 2010 level***
Subtract:	***---------- -***
	Level in 2010

Figure 1a illustrates the adaptation. The model first calculates the reduction in ozone levels between 1990 and 2010 for each grid cell (5% in the example). Then, for each available monitoring station within the grid cell, this reduction is applied to the 1990 measurements to find the ozone level in 2010.

The results of the method can be regarded as simulated measurement results for 2010 per station. In grid cells where the network density is similar to the future 2010 network, the method yields an approximation of the measured results in 2010. A disadvantage of the method is that where there are no measurement stations it gives no results.

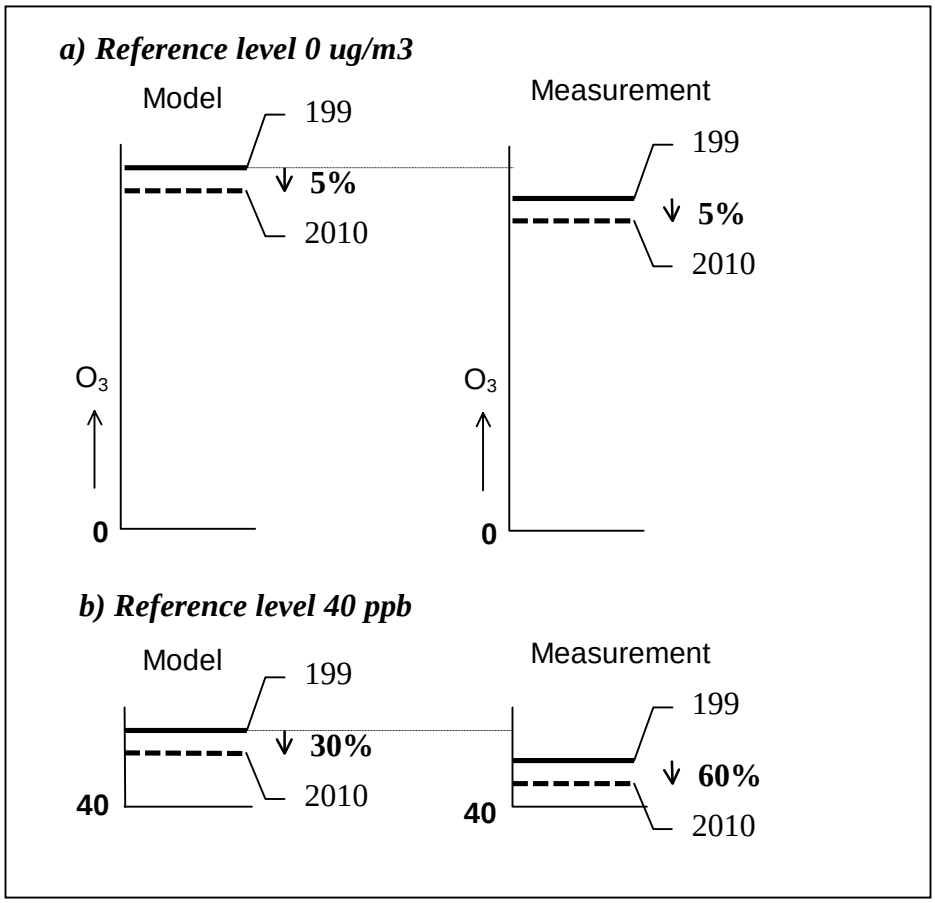

Figure 1. (a) Illustration of the adaptation of model results to measurements; (b) Dependence of the reduction percentage on the reference level

Application of modelled reduction between 1990 and 2010

To apply the method, it has to be specified how the reduction in ozone between 1990 and 2010 must be defined and applied to the measurement results. The simplest approach would be to determine the modelled percentage reduction in the target value parameters AOT40 and NET60[25] and apply this to the measured values of AOT40 and NET60. This is, however, not correct, as Figure 1 illustrates. In Figure 1a a calculated 5% reduction of a certain hourly concentration due to emission reduction is applied as 5% of the corresponding measured concentration. If, however, this reduction is expressed as percentages of the *exceedance* of a certain level, as in Figure 1b, the percentages are no longer equal. This applies to both AOT and NET, which are defined relative to a reference level.

Consequently, it is not possible to derive directly from the reduction percentage calculated by the model for NET or AOT a reduction that can be applied to measured values. We need to apply the reductions calculated for the total concentrations (reference level zero) and apply these to total measured concentrations and then derive NET and AOT from the results.

An option is to use the model predictions of the reduction percentage for each individual hour (the time series) and apply the reduction percentages calculated by the model per individual hour to the

[25] Number of Exceedance in days of the Threshold of 60 ppb (= 120 µg/m³), the health related target value)

measured level of each hour[26]. The resulting time series for 2010 can be used to calculate the value of NET and AOT in 2010.

An intermediate approach could be more effective, in which we use not the reductions predicted for the primary output of the EMEP/RAINS model (i.e. the time series), but the reductions for an aggregate of EMEP/RAINS model results. This could be done by constructing the frequency distribution of the time series, determining the reduction percentage as a function of the percentile and applying this to the measured frequency distribution.

Both methods were applied in the analysis:

Time Series (TS) method

Apply the reduction percentages calculated by EMEP/RAINS for each individual hour of the time series to the corresponding measured concentration of that hour and calculate for the predicted time series the new NET60 and AOT40.

Frequency Distribution (FD) method

Sort the calculated and measured hourly concentrations separately according to magnitude. This yields the frequency distribution of the concentrations. Apply the reduction percentages calculated by EMEP/RAINS for each individual percentile to the measured concentration percentile and calculate from the resulting frequency distribution the new NET60 and AOT40.

Measurement results used

Measured data were used for the "base case" years around 1990 that were taken in the EMEP/IIASA model calculations: 1989, 1990, 1992, 1993 and 1994. The data sets for more recent years are larger and probably of better quality, but there were no scenario calculations that could be related to later years. The model results pertained only to the period April-September, so the use of the measured data was also limited to that period.

To obtain an impression of the dependence of results on the selection of the stations, two sets of station data were taken for the analysis:

1. The **EMEP Stations** in EU countries (48 rural stations);

2. The **Selected Stations**, a set of stations in Austria, Germany, Belgium and the Netherlands (in total 46 stations for 1990-1994; for 1989, German data were not available because of possible calibration problems, leaving 26 stations in the other countries). The stations were selected by Austria, Germany and Belgium on the basis of a request to provide data from rural stations that existed during the five years with a high data capture. For the Netherlands TNO selected the stations.

The EMEP Stations set has the advantage that it gives to some extent an overview of the EU situation, though coverage is far from complete. The Selected Stations set was added to obtain more detailed information on the regions where the highest measured concentrations were found (the Benelux/Germany area and Austria with its mountainous stations). Stations from southern Europe and France are lacking in both sets, since these were unavailable, or available only for a limited period during the base case years.

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[26] Since the EMEP model calculates hourly concentrations only at 6h intervals, the reduction percentages have to be interpolated to hours in between.

Results

Four sets of results emerge from the calculations: results of the TS and the FD method for each of the two sets of stations. Instead of giving maps, the results are expressed in terms of the frequency distribution over the set of stations. In section 6 of this Annex it is explained that the highest value of all stations is not useful for guiding the derivation of the target value, but that the 90 percentile is taken for this. The 90 percentile of AOT40 represents the value that is exceeded at 10% of the stations[27].

The most relevant form for the results is that of the proposed Target Values, viz a three-year average for NET60 (and for completeness AOT60 as well) and a five-year average for AOT40. Table 1 presents the 90 percentile of these averages for the two sets of stations and the two method TS and FD. For comparison the results of the EMEP/IIASA model calculations are also shown.

Table 1: Mean of 3 years for NET60 (days) and mean of 5 years for AOT40 (ppb.h): 90 percentile of stations

a EMEP stations

		EMEP/RAINS model		Measured	TS method	FD method
		1990 emissions	2010 emissions		2010 emissions	2010 emissions
NET60	89-92 [1]	40	14	41	*32*	*15*
NET60	90-93 [1]	37	13	39	*29*	*15*
NET60	92-94	33	13	42	*32*	*15*
AOT40	89-94 [1]	15132	8634	10354	*8539*	*5075*

b Selected stations

		EMEP/RAINS model		Measured	TS method	FD method
		1990 emissions	2010 emissions		2010 emissions	2010 emissions
NET60	89-92 1	45	23	48	*48*	*23*
NET60	90-93 1	45	18	51	*43*	*20*
NET60	92-94	43	18	55	*46*	*23*
AOT40	89-94 1	15648	10874	11492	*10309*	*6654*

[1] Excluding 1991

[27] One should bear the limited size of the data set in mind: e.g. for results from 45 stations the 90 percentile corresponds approximately to the fifth highest value.

Discussion

The calculated 90 percentiles for the base case years are not very far away from the corresponding values from measurements, particularly in view of the strong inherent variability of the NET and AOT parameters. The interannual variability range is in many cases larger than the mismatch between model and measurement. The agreement for individual stations is less good. This is not surprising: non-systematic model inaccuracies and measurement errors have more effect on the results of individual stations than on the overall frequency distribution. One should therefore be cautious in the interpretation: the stations corresponding to the 90-percentile are generally not at the same location as the grid cell corresponding to this percentile in the model results.

In trying to interpret the percentiles of the station distributions one should be aware that the modelled "hot spot" region (Benelux and surroundings) does not fully coincide with the area where the highest measured levels are found.

The FD method reproduces the modelled reductions reasonably well, whilst for the TS method the reductions in NET60 and AOT40 are generally found to be much less than the modelled reductions. It is not *a priori* clear which of the two methods gives the more reliable results. The tendency of the TS method to give much lower reductions than the model can be understood as follows. The EMEP/RAINS model has a strong tendency to link the largest reduction percentages to the highest concentrations. Since model and measurement results are not perfectly correlated, the method links the largest reduction percentages in many cases to measured concentrations that are not high, and, conversely, some of the highest measured concentrations are linked to lower reduction percentages or even negative reductions (increases). So, the TS method can be expected to systematically underestimate the reductions.

It is less clear whether the FD method would tend to overestimate or underestimate the reductions. Obviously, the EMEP/RAINS model tends to link the highest reduction rates to the highest levels, and so the FD method tends to link the high reduction rates to the highest concentrations, even if they are measured on a different day with different meteorological conditions. It is difficult to assess how important this is. The assumption that the highest levels are more sensitive to EU emissions reductions than average levels is generally plausible, but not all occurrences of high levels are due to EU emissions; stratospheric intrusions are a clear example. It is not to be expected that in reality high ozone days would be more strongly correlated with the high reduction rates than the model predicts. On the other hand, when high concentrations are measured on days for which the meteorological conditions cause the model to predict a low sensitivity to emissions, it is quite conceivable that the low response to emission reductions is realistic. So, although the modelled dependence of the decrease of NET60 and AOT40 on the 1990 ozone level is obviously much better approximated by the FD method than the TS method, the FD method probably tends to overestimate the reductions.

The exercise may therefore be regarded as a sensitivity analysis which indicates the expected range of predicted levels, and in which the FD results may be the most plausible.

Proposal for the target values

In deriving a target value also involving measured concentrations, one should avoid the following sources of uncertainty and inconsistency:

(a) Extreme values which are often susceptible to measuring and modelling errors;

(b) Peculiar local scale features of ozone formation are likely to encourage very high concentrations. Such should not determine the setting of the target value in order to maintain its role of as a benchmark for the EU abatement strategy, which necessarily focuses on a regional scale.

In order to exclude these factors a 90 percentile of the available stations in each set was taken rather than the absolute maximum. So, 10% of the series, i.e. basically 5 stations in each set of EMEP and 'selected' stations were still allowed to exceed a possible level for the target value.

NET60: Table 1 presents the 90 percentile of the numbers of exceedances for 2010 for the three combinations of consecutive meteorological years[28]. The reference data are also given, viz the exceedances predicted by the model for 1990 and 2010 and the measured values. For the TS method, the value of NET60 predicted for 2010 is only about 8 (0-10) exceedance days (about 20%) lower than the measured value for the base case. For the perhaps more realistic FD method, the reduction found agrees well with the modelled reduction: 25-32 exceedance days (about 60%). The number of exceedance days remaining is according to the FD method 10-14 for the EMEP Stations set and 20-22 for the Selected Stations set. If it is assumed that the FD method provides the most realistic results and that 1989 is high due to measuring errors, a target value of NET60 of the order of 20 is obtained.

AOT40: Again basing the target value on the 90 percentile of stations, a value of about 8 ppm.h (TS method) or 5 ppm.h (FD method) is found for the EMEP stations. For the Selected Stations the values are 10 and 7 respectively.

Limitations and Uncertainties

Model data:

At the time of writing, IIASA's 7th Interim Report was available. The results presented in Table 1 are based on that report and pertain to the H1 scenario.

Measurement data

Although the data from both sets of stations had passed the quality control procedure of the network, doubts on some stations arose during the processing of the data. In contacts with the network personnel these doubts were sometimes confirmed. Especially in 1989 calibration problems seem to have existed for several stations. Some of the stations exist in both sets and for several of these the data sets received (from EMEP and Member States respectively) were not identical. Since it was not possible to identify erroneous results in a systematic way, a selection of data that should be removed could not be made. These limitations should be borne in mind in the interpretation of the results.

Analysis

The analysis described above has some deficiencies worth noting. Several of these can be ascribed to the limited time available for the analysis. However, even an extended analysis would suffer from uncertainties that are insoluble. It is not realistic to expect that the quality problems for the measurement data around 1990 can be solved by more detailed analysis. The coverage of the EU territory can be improved by extending the data set with e.g. data from France, but the fact will remain that there are no reliable data for many areas. Only when the base case period from 1990 is changed to a more recent year can a large step forward in the availability of reliable monitoring data be expected. The large uncertainty associated with the difference between the TS and FD method cannot be reduced by improved analysis because this uncertainty results primarily from the limited correlation between the modelled and measured concentrations. Also the uncertainties due to the limited spatial correlation between model and measurements are independent of this analysis.

[28] Other combinations might be taken also; 1992 is part of all three combinations taken here.

Finally, the results are sensitive to the choice of the percentile of the stations sets, the substantiation of which cannot be improved by more detailed analysis of the method.

Consequently, it is not expected that a more extended analysis will significantly improve the basis for choosing the levels of the target values.

Conclusion

The adaptation of model results to measurements has considerable uncertainties. A point of concern is that the locations where the EMEP/RAINS model finds the highest levels only partly coincide with the locations found in measurements. Some of the high measured levels at such locations seem to be unreliable, and it is not possible to quantitatively assess the importance of measurement errors in general. Several elements of the method could be improved, but the most important uncertainties are expected to remain. The two methods applied, TS and FD, respectively give a much smaller decrease between 1990 and 2010 than the model predicts and a comparable decrease. Assuming that the FD method gives the more plausible results and omitting the highest 10% of the stations because of inaccuracies or local causes, a value of around 15 exceedance days is found for the EMEP station set and around 20 for the Selected Stations set. One should be aware that uncertainties due to limited representativeness of the EMEP network and of the 5 years chosen, the much higher results of the TS method, etc. tend to increase these values.

Acknowledgement

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ANNEX D: THE EFFECT OF LOCAL AND REGIONAL EMISSION REDUCTIONS

Introduction

Chapter 5.1 considered the impact of emission reductions on ozone levels, as viewed from the EU-wide perspective. The integrated assessment model RAINS allowed systematic exploration of various strategies, even in quantitative terms of air quality objectives and costs. Unfortunately, no similar approach yet exists for developing strategies to reduce ozone at the urban and regional scale. Of course, emission reductions resulting from the EU-wide strategy will also affect urban and regional[29] emissions, but the RAINS calculations do not address the way in which local emission reductions relate to local ozone levels.

This is not due to scientific neglect of the issue. Indeed, numerous investigations of the relation between urban and regional emissions and ozone levels have been reported. The picture emerging from these studies is, however, far from clear. This is due not so much to conflicting scientific results, as to the widely varying conditions to which the results pertain. In particular, local depletion of ozone near sources, due to rapid titration by freshly emitted NO, in combination with slower formation of ozone by NO_x and VOCs makes it difficult to form a generic picture in quantitative terms. The balance between ozone destruction and formation depends on: the size of the area in question, the solar radiation governing the photochemical formation rate, the wind fields governing the residence time of the polluted air mass, and the composition of the precursor mixture both in emissions and in upwind air. All these conditions vary across Europe with climatic and topographic characteristics, as well as through the year with weather changes.

Studies on local and regional scale reductions

Numerous studies with relevance to the subject of this Annex have been conducted. This section gives an impression of the results, but is certainly far from complete.

Experimental and modelling studies in Germany

In order to improve understanding of the effectiveness of local short-term measures, several large studies have been undertaken in Germany, encompassing field measurements and model calculations. Also, experiments in Heilbronn and Hessen were conducted in which temporary emission reduction measures were applied during an ozone episode. A German working group summarised the results and tried to draw conclusions (Bruckmann and Wiechmann-Fiebig, 1997). The conclusions pertain to short-term reductions during episodes, but also provide insight into the effectiveness of durable measures, though the types of measures are generally different. The conclusions drawn for the situation in Germany include the following:

- Only large-scale (western and central Europe) and permanent reductions of precursor emissions will solve the problem of elevated ozone levels.

- Downwind of large conurbations, distinct ozone plumes were observed with peak concentrations exceeding background levels by 15-40%. This additional burden can be

[29] The term 'regional' refers to areas with dimensions of up to a few hundred kilometres.

reduced by short-term regional actions. These cases are however rare events (several days in nice summers).

- Regional, temporary abatement actions can effectively reduce (10%) ozone peak concentrations in urban plumes provided that:

    * measures include at least one big conurbation with several million inhabitants (or a corresponding emission flux);

    * abatement actions reduce precursor emissions more than 20%. Slight regional reductions of NO_x emissions (<10%) do not lower ozone peaks significantly.

- Because of the non-linearity of the ozone-precursor relationship, ozone maxima can be reduced by no more than 10-15% if precursor emissions are reduced by 30-40%. The size and duration of threshold exceedances in the ozone plume can be more than halved by these emission reductions under favourable conditions.

- Traffic bans in big conurbations or regions reduce NO_x and VOC emissions by nearly 30%. Speed limits (90/80 km/h) alone are not sufficient. Speed limits (80/60 km/h) which cover Germany as a whole can reduce NO_x emissions by more than 10% and can serve as an additional measure.

- Regional traffic bans should be supplemented by abatement actions in the industrial and solvent use sector. Corresponding measures are, however, very difficult to enforce.

South European studies

Annex B considers the particularities of ozone in south European regions. Compared with more northerly parts of Europe, topographic influences in the south tend to keep air masses above conurbations more isolated from each other, so that local emission reductions tend to be more important. Some big cities are known for their air pollution problems, e.g. Athens and Milan. In some of these cities ozone formation has been extensively studied, and insight into the effectiveness of reduction measures is gradually growing. A generalised picture is, however, still far away.

Numerous studies have been published about Athens, e.g. the MEDCAPHOT-TRACE experiment (MEDCAPHOT-TRACE, 1998) which focused on experimental campaigns to understand ozone, but also addressed the effectiveness of reduction measures. From model calculations it was concluded that the ozone abatement strategy should focus primarily on VOC emission controls rather than NO_x emissions controls, since NO_x control tended to increase ozone. Traffic reduction measures alone, which involve both NO_x and VOC reductions, seemed to have no net effect on the maximum ozone levels observed.

An example of a study on the Milan metropolitan area is the application of the CALGRID-CALMET modelling system for the evaluation and comparison of different emission control strategies (Finzi *et al*, 1998). Since vehicular traffic is the main source of pollutants in the area, the focus was on the impact of cleaner fuels and cleaner cars. In the simulations it was found that the use of compressed natural gas in light vehicles or the introduction of electric cars in the city centre would result in an appreciable reduction of VOC and ozone, while oxygenated fuels did not seem to be significantly effective; also, catalytic converters had a downward effect on ozone.

For the Valencia region, described as an example in Annex B, Cuvelier and Thunis (1998) carried out model simulations of regional precursor reductions corresponding to the REF scenario (see Chapter 5) and the more stringent so-called Maximum Feasible Reduction scenario. Assuming background levels to remain unchanged, they found that ozone tended to increase in the coastal area, where ozone was relatively low, while in the mountains, away from sources, it tended to go down.

Other plume studies

Studies of ozone 'plumes' of cities and regions give insight into the local contribution to downwind ozone levels. Results of plume studies in Germany (notably the FluMOB project in the Berlin/Brandenburg region (Stark and Lutz, 1997) and plume measurements near Munich and several cities in Saxony-Anhalt) are already covered in the conclusions of the German studies mentioned above. Other work is mentioned in this section.

In Austria the Pannonische Ozon-Project (POP) (Baumann *et al*, 1997) was conducted, in which both measurements and model calculations were carried out. One conclusion of the study is that typical local production in the plumes of the city of Vienna might account for close to 25% of the total ozone level. Reduction scenario simulations showed that the response of ozone to national emission reductions depends very much on the situation (air mass trajectory, NO_x/VOC ratio). No sub-national emission reductions were investigated. Another large programme (POLLUMET) was conducted in Switzerland, and included ozone plume measurements. Relative differences of the ozone concentrations were typically around 30 %, while downwind of big cities elevated ozone values of up to 70% above upwind levels were recorded (BUWAL, 1996).

Auto-Oil I

In the first Auto-Oil Programme both large-scale and urban-scale reductions were studied. For the urban scale two separate modelling studies were conducted: calculations with the IVL Lagrangian chemical model (Simpson and Andersson-Sköld , 1996) and with the more detailed 3-dimensional EZM model (Moussiopoulos *et al*, 1996).

The calculations with the IVL model were done for six model cities in the EU. A large number of hypothetical ozone episodes were studied, in an attempt to obtain a reasonably statistical picture. Partly due to the statistical nature of this approach, the results are more qualitative rather than quantitative with respect to the effects of emissions. The more rigorous calculations by the EZM model could be considered more reliable, but here the drawback was that only three episodes could be studied: two for Athens and one for Cologne.

For background levels in 2010, the results of EU-wide reduction scenario calculations were taken. Both studies predicted that an additional 50% reduction of local NO_x emissions alone could have adverse consequences for ozone in the cities (though for one episode in Athens ozone was found to decrease). VOC emission controls at least as stringent as those for NO_x, and possibly more stringent, seemed to be required to overcome the adverse effect of NO_x on urban ozone.

OFIS study

As a first step towards a statistical overview of urban ozone formation, the European Commission commissioned a modelling study (Moussiopoulos, *et al*, 1998) to develop statistics on ozone formation and its response to urban emission reductions for two example cities. In this study a simplified urban photochemical model, OFIS, was applied to Stuttgart

and Athens. Emission reductions and air pollution levels resulting from the EU-wide scenario calculations of the EMEP/RAINS model (see Chapter 5) were taken as starting points for examining the effect of further local emission reductions. The simplifications in the model made it possible to carry out simulations for each day over a six-month period (1 April to 30 September 1990). Comparison with available measurements showed a satisfactory agreement for Stuttgart; the model performance was less satisfactory in the case of Athens, as the model did not resolve local circulation systems.

The extended period made it possible to express the results in statistical terms, including NET and AOT values. Lower ozone burdens were found in the case of one-sided VOC emission reductions. One-sided NO_x emission reductions on the other hand seemed to lead to more ozone in the urban area, while benefits could occur at fairly large downwind distances. The calculations suggested that additional local NO_x emission reductions of the order of 70-80% would be required for an acceptable reduction of the overall ozone burden. It was concluded that in Central European airsheds like Stuttgart it is very important to combine emission reduction measures at both the regional and the urban scale. To a lesser extent this conclusion was deemed to be valid for isolated areas like Athens, where local measures have a larger share in reducing the urban ozone load.

Further application of OFIS to 23 urban areas, carried out in the framework of other European studies, confirmed that the results for the city of Stuttgart might be transferred to the majority of EU urban areas.

USA experience[30]

In the USA ozone has been recognised as a serious issue for several decades. This early recognition was mainly due to the very high ozone levels in the Los Angeles basin. Ozone abatement strategies were already implemented around 1970 and focused mainly on VOC reductions.

Though this strategy brought about a decrease in peak ozone levels, many areas still failed to meet the standards by the end of the eighties, and in some areas ozone had even increased. An extensive assessment of this disappointing outcome resulted in the report "Rethinking the ozone problem in urban and regional air pollution"[31]. The report highlighted the role of several uncertainties, in particular the confounding effect of meteorological fluctuations on ozone trend detection and the lack of reliable methods to monitor emission reductions. There were indications that the VOC emission data were too low by a factor of two. As NO_x reductions are more effective when the NO_x/VOC ratio is higher, this raised doubts regarding the focus on VOC reductions. Added to this was the indication that in some areas the impact of biogenic VOC emissions was underestimated, which again tended to push NO_x reductions higher up the agenda.

Following this report several field campaigns were set up, with three basic tasks: to study ozone formation, to assess whether ozone in the area of study was VOC or NO_x controlled and to validate models. In 1995 the research programme 'North American Research Strategy for

[30] Based on P.J.H. Builtjes (1998). Overview and analysis of international activities in the field of photo-oxidant modelling. TNO-MEP report R 98/164, Apeldoorn, the Netherlands.

[31] NRC (1991). Rethinking the ozone problem in urban and regional air pollution.

Tropospheric Ozone' (NARSTO) was established to promote the development of strategies for local and regional ozone management.

Final conclusions have yet to be drawn from these studies. The impression is that no major flaws in the models have been detected and that the effectiveness of NO_x versus VOC control depends on the area under consideration. It is very noteworthy that US ozone strategies focus mainly on the urban and regional scale; the issue of long-range transport receives less attention. For the north-eastern part of the USA, the Ozone Transport Assessment Group (OTAG), set up by the EPA, has recommended strategies for reducing ozone transport, and these form the basis for the development of legislation on interstate ozone transport.

Conclusions

A comprehensive analysis of the effect of local and regional-scale emission reductions on ambient ozone levels (such as the RAINS model calculations provided for large-scale ozone) is not yet possible. In experimental studies it is difficult to unambiguously distinguish city plumes from spatial fluctuations in the background levels, and the number of studies is too small for rigorous generalisation. Model studies on the other hand have in principle more potential for generalisation, but inaccuracies in emissions, dispersion and chemistry, along with compromises needed to overcome computing time problems, render the results uncertain. Conclusions therefore have to be drawn in qualitative terms, based on several studies in Europe and on information from the USA.

In the USA a simple generalised picture has not emerged, despite considerable concern over local and regional ozone and a long history of research in that area. Also, one should be wary of extrapolating the results to Europe. US cities are typically larger than their European counterparts, and meteorological conditions (30-35°) ideal for local ozone formation. Biogenic emissions are also expected to have a much greater impact on ozone formation in the USA than in most of Europe. The differences between the USA's focus on local and regional strategies and the European emphasis on long-range transport have not been fully analysed, however.

The general picture is that within and very near to source complexes like cities ozone is depressed and that reduction of local emissions tends to increase local ozone. At sufficiently large distances from the sources, emission reductions generally cause ozone levels to go down. The distance where net depletion turns into net formation depends very much on the situation: it depends on the climatological conditions of the region concerned, on the particular episode that is studied, on the size of the area in which emissions are reduced, on the NO_x/VOC ratio in emissions and in upwind air, on the reactivity of the VOCs emitted and also on the existence of downwind sources. The complexity results to a large extent from the fact that NO_x reductions can both reduce and increase ozone. Anthropogenic VOC reductions on the other hand virtually always reduce ozone, though the size of the ozone reduction depends very much on the local conditions. On the local and regional scale in particular, the effectiveness of VOC reductions depends to a large extent on the reactivity of the VOC species.

To this general picture some tendencies can be added which all increase the probability that NO_x reductions will lead to ozone reduction:

- During episodes of high ozone, photochemical formation is strongly enhanced, which makes the counter-effective titration of ozone by local NO emissions relatively less important.

- When the emission reduction area is increased, the average residence time of the air in the area increases, giving more time for ozone formation to compensate the depletion.

- During episodes of low wind speeds and in areas where topographic influences impede refreshment of the local air mass, the increased residence time of the air mass gives more time for ozone formation.

- In southern Europe the residence times of air masses are generally larger, and photochemical reactions faster, than in more northerly parts of the EU, favouring the ozone formation process.

- In areas of low NO_x/VOC ratios in emissions and concentrations, the balance of depletion and formation by NO_x tends more towards formation, which makes NO_x reduction more effective for reducing ozone.

- Downwind of a single source area like a city ambient NO_x is lower than in regions where NO_x emissions are nearby everywhere, and so the depletion at ambient level of ozone by NO_x tends to be less important.

Clearly, not all tendencies are independent. Table 1 attempts to summarise how local ozone levels respond to local emission changes. It is important to note that the table pertains to ozone changes resulting from emission reductions that occur only locally; when concurrent large-scale reductions are undertaken, the total net effectiveness of NO_x reductions increases.

Table 1 Indication of the response of high ozone levels to local reductions of precursors

Location of ozone levels	Only local NO_x emissions ↓	Only local VOC emissions ↓
	Local ozone change during episodes	
City centres	mostly ↑ *	↓
City surroundings	↓ or ↑ *	↓
Regional rural areas	↓ or ↑ *	↓

* The probability of ↓ tends to increase when:

- the ozone levels are higher;
- the emission reduction area is larger;
- the residence time of air masses is shorter;
- the location is more southerly;
- the NO_x/VOC ratio in local emissions and concentrations is lower;
- ozone is measured further downwind of single source area.

For better insight, statistical information is needed on the response of ozone to emission changes. In the first place, results should pertain not only to individual episodes, but to an ensemble of episodes. The most relevant statistics are NET and AOT values on an annual basis. In addition, general information on the effectiveness of NO_x and VOC control is needed, as a function of local and regional climatological conditions, source composition, VOC reactivity and receptor location relative to the sources. Such generalised information is currently lacking. A first attempt to develop such statistics was undertaken in the OFIS study, but it used a simplified model; more detailed models can be employed to give more accurate data, though it is difficult to estimate how accurately the borderline between net ozone depletion and net formation can be calculated.

The above conclusions pertain to both durable and short-term measures; during an episode, ozone reductions do not depend on whether local precursor emissions reductions are due to short-term or durable measures, provided that short-term measures have been implemented

soon enough. Short-term measures, however, could be restricted to weather conditions during which local scale reductions are most effective.

For durable measures one cannot make this differentiation, since they will be in effect throughout the year[32]. One has then to judge the effectiveness of ozone reductions in terms of long-term statistics of high ozone levels, in particular NET and AOT. Such statistic might reveal as if the effect of durable measures was small compared to short-term actions, because durable measures will also be in effect when they are less effective.. However, in many cases short-term measures would be initiated too late and over too limited an area to be effective in preventing elevated ozone levels.

It is also difficult to assess cost-effectiveness, as no studies have yet been performed evaluating real cost of implementing short-term measures and comparing it with the benefits. While some short-term emission reductions can have a more disruptive effect on economy and society than durable ones others might be quite cheap to implement

[32] An intermediate type is the seasonal emission reduction, which is not triggered by high ozone levels, but is in effect during the season in which episodes are expected.

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ANNEX E: URBAN AND REGIONAL SCALE MEASURES

This Annex provides a summary of measures that could be taken at urban or regional level, in addition to national or international measures. The measures are divided into durable measures, of a long-term nature, and short-term measures, which are only taken during or in anticipation of periods of high ozone levels. The list contains not only legally enforced actions to reduce emissions, but also more indirect actions, e.g. awareness raising, which may also lead to emission and/or risk reduction.

The list has been compiled from various reports and publications of local measures, including the OFIS study mentioned in the main body of Chapter 5.3 and an action guide by ICLEI. It is not intended to be exhaustive; many measures at national scale could also be taken at the regional or local scale; for the industrial/commercial sector in particular, the list might be considerably expanded. Also the distinction between durable and short-term measures should not be viewed as absolute: some durable measures can be implemented for a short period or during the smog season, whilst some short-term measures may also be applied as durable measures.

At the end of this Annex a summary is given of national legislation on short-term measures.

Durable measures

General information and appeals:

- General information on ambient ozone in brochures, leaflets, teletext, internet, (free) telephone, etc.
- Information on current and forecast ozone concentrations on teletext, internet, etc.
- Information for general practitioners regarding ambient ozone and effects
- Public display of information (ozone levels, recommendation on exposure behaviour)
- Information on concentration levels, aimed especially at sensitive groups
- Signs and slogans on buses
- Encouragement to report smoky diesel engines

Traffic-related measures:

- Stimulate shift towards public transport, bicycle, foot
- Traffic flow planning
- Areas barred to private vehicles
- Road pricing
- Integration of transport management and air quality control
- Road signing and traffic signalling
- Promotion of car-pooling
- Park-and-ride facilities
- Parking area management
- Subsidise scrapping of old vehicles
- Stage II at petrol stations (if not a measure at EU level)
- Faster implementation of EURO IV standards for passenger cars and light duty vehicles
- Full implementation of emission standards for mopeds and motorcycles
- Facilities for commuters to combine bicycle and public transport
- Plan houses closer to work, spare time activities and shops
- Spatial planning according to public transport
- Stimulate reduced engine power

- Co-ordination of goods transport by different companies; central city distribution system
- Incentives to rail cargo
- Traffic-calming provisions
- Bus lanes
- Remote sensing control of motor vehicle emissions
- Incentives to low emission buses and trucks

Household-related measures:

- Promote water-based paint (in addition to possible EU measures)
- Promote low-NO_x boilers and water heaters
- Substitution of solvent content in household products (cleaning, sprays, detergents etc.)

- Promotion of renewable energy and improvement of energy efficiency

Industry/commerce-related measures:

- Stricter demands for VOC and NO_x emissions in permits
- Intensify enforcement of air pollution legislation
- Emission reduction from construction sites
- Emission reductions from cleaning processes
- Emission reductions from surface coating processes
- Closed systems for organic liquid storage and distribution
- Promote natural gas penetration
- Government security for emission reduction investments of small industries
- EIA for land use
- Energy-saving programme for buildings

Short-term measures

General information and appeals:

- Information when high levels are occurring or expected, in addition to the information in the framework of the ozone directive.
- Appeal to the public to reduce polluting activities
- Appeal to businesses to reduce polluting activities
- Interviews on TV
- Recommendation to turn engines off in the event of long stops

Traffic-related measures:

- Banning certain types of traffic (partial or complete)
- Banning the most polluting passenger cars
- Speed limits
- Restriction of traffic in inner city (e.g. certain hours of the day, permit schemes)
- Promote cycling, car-pooling, public transport use
- Reduce the price of public transport tickets
- Introduce temporary parking restrictions in inner city

Household-related measures:

- Restrict VOC-based paint use

Industry/commerce-related measures:

- Switch to other fuel
- Restrict VOC and/or emitting activities
- Ban certain VOC and/or emitting activities

Legislation on short-term measures

In a recent inventory of legislation on short-term measures (LFU, 1998) it was found that in the countries investigated (viz. EU and USA) statutory short-term measures to reduce ozone precursors exist in France, Greece, Austria and the USA. In all cases the responsible authorities are at regional or urban level.

- France mentions in its national air pollution law of 20 December 1996 alert values of 360 μg/m³ for ozone, 400 μg/m³ for NO_2 and 600 μg/m³ for SO_2, which trigger short term measures. The measures include driving restrictions for vehicles that are relatively polluting. Cleaner vehicles are marked by a plaquette.

- In Greece short-term measures only exist at the local level for Athens. The trigger for such measures is not ozone, but NO_2. When 500 μg/m³ is exceeded, alternating driving bans come into force for odd and even licence numbers in turn.

- Austria has two alert levels of 300 μg/m³ and 400 μg/m³ in its ozone law of 24 April 1992, which trigger short-term measures, including driving restrictions.

- In the USA the air pollution level is indicated by the Pollution Standard Index (PSI), which is composed of the levels of several pollutants. When 100 points are reached information is given to the public. In summer this is the case at exceedance of 240 μg/m³ ozone. This public information includes appeals to car drivers to abstain from driving or to pool cars. There are no statutory driving bans.

References

ICLEI. (1996). European Cities and Ground-level Ozone. A Guide to Regional and Municipal Action Strategies. The International Council for Local Environmental Initiatives. European Secretariat GmbH, Freiburg, Germany.

LFU (1998). Kurzzeitmassnahmen zur Reduziering der Ozonevorläuferstoffe in der Europäischen Union und den USA (1998). Landesanstalt für Umweltschutz Baden-Württemberg, Karlsruhe, Germany.

ANNEX F: INFORMATION TO BE RELEASED TO THE PUBLIC IN THE EVENT OF EXCEEDANCE OF THE INFORMATION THRESHOLD OR THE GENERAL ALERT THRESHOLD

For reasons of harmonisation it would be desirable for Member States, in the event of exceedance of the information threshold or the general alert threshold, to issue exactly the same information to the public. Since, however, the methods of providing the information and the contexts may differ between Member States, a certain degree of flexibility is preferred. It is therefore proposed that the Directive include only the minimum information that should be given to the public.

Minimum information to be issued when the information or general alert threshold is exceeded or expected to be exceeded

In the event of exceedance or expected exceedance of the information or general alert threshold, the text released to the public must contain at least the following elements:

(1) Information on observed exceedance(s):

- Location or area of the exceedance;
- Type of threshold exceeded (information or general alert);
- Time and duration of the exceedance;
- Highest 1-hour and 8-hour mean concentration.

(2) Forecast for the following afternoon/day(s):

- Time period and geographical area of expected exceedances of information and/or general alert threshold;
- Predicted 1-hour maximum concentration or range of concentration;
- Expected change of pollution (improvement, stabilisation, or deterioration);
- Reason for occurrence and/or expected change in the situation.

(3) Information on type of population concerned, possible health effects and recommended conduct:

- Information on groups of population at risk;
- Description of likely symptoms;
- Recommendations on precautions to be taken by the population concerned;
- Where to find further information.

(4) Information on preventive actions to reduce pollution:

Indication of main source sectors; recommendations for action to reduce emissions