

Position paper on Air Quality: nitrogen dioxide

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NOTE

This document reflects the opinions of the majority of the members of the Working Group.

It should not be considered as an official statement of the position of the European Commission

Not all experts necessarily share all the views expressed in this document.

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Preface

At the meeting of National Experts on Air Quality Group, 5 July 1995, four working groups were established for preparation of position papers for the development of daughter legislation required under the draft Council Directive on Ambient Air Quality Assessment and Management. The Directive was adopted by Council on 27 September 1996 (96/62/EC). It is referred to in this document as the Framework Directive (FWD).

This position paper has been developed for NO₂ by a working group lead by Denmark and Sweden with participants from Austria, Belgium, Finland, Italy, Commission's Joint Research Centre (JRC), European Environment Agency (EEA), industry, NGOs and DGXI. Representatives from the World Health Organisation (WHO) were invited to participate when relevant.

This position paper has been prepared in accordance with the recommendations from the Commission (AAQ/95/1/2) including the amendments from the meeting on 5 July 1995.

The Working Group (WG) has had four meetings. The first meeting was held in Copenhagen on the 12 and 13 October 1995 and the main topic was preparation of the Risk Assessment chapter of this position paper. The second meeting was held at the Joint Research Centre at Ispra, on the 11 and 12 January 1996. The main topics of the second meeting were measurements, assessments and quality assurance. The third meeting was held at Swedish Environmental Research Institute (IVL) in Gothenburg, on the 25 and 26 March 1996 with the main topic being cost implications. The fourth and last meeting for finishing the work of the group and the position paper, was held in Vienna 3 - 4 December 1996.

The available data indicate that at least 21 million people in Europe are exposed at times to daily NO₂ concentrations above the WHO 1987 Air Quality Guideline(AQG) of 150 µg·m⁻³ for 24 hours. If these data represent also the air quality in cities where no measurements have been made, 20% of the European urban population are exposed to levels exceeding the WHO 24 hour AQG (1987).

Scope

This paper has been prepared in accordance with the Directive 96/62/EC on Ambient Air Quality Assessment and Management and DGXI guidance as outlined in document AAQ/95/1/2. It addresses meeting NO₂ air quality objectives for the protection of human health, ecosystems and the environment, risk assessment, present and future air quality and the costs and benefit of setting objectives. It does not address indoor air pollution, deposition or synergistic effects between NO₂ and other pollutants, or climatic effects of nitrogen-containing pollutants. As per the Annex I of the Directive it focuses on NO₂.

In determining limit values and, as appropriate, alert thresholds for NO_2 the following elements must be kept in mind:

- Abatement strategies for reducing effects of NO_x may in some areas, lead to increasing photochemical ozone concentrations.
- Parts of the population may be exposed to significant indoor and/or workplace NO_2 pollution, but this is outside the scope of the Directive.

1. Introduction

1.1 Nitrogen compounds in the air

Nitrogen compounds are present in the atmosphere in both oxidised and reduced forms. The reduced compounds include ammonia (NH_3) and ammonium (NH_4). The oxidised compounds include nitrous oxide (N_2O), nitrogen oxide (NO), nitrogen dioxide (NO_2), nitrous acid (HNO_2), nitric acid (HNO_3), peroxyacetylnitrate (PAN) and particulate nitrate (NO_3^-). NO_x is defined as $\text{NO} + \text{NO}_2$.

Nitrous oxide (N_2O) is emitted to the atmosphere due to bacterial activity in the soil. It is emitted also from anthropogenic sources (e. g. catalytic reduction processes) and acts as a greenhouse gas. However, it is disconnected from the NO_x chemistry in the atmosphere and is not important for assessment of air quality.

Nitrogen oxides are emitted mainly (in most cases >90%) as NO . NO_2 is formed relatively rapidly from NO by reaction with ozone or radicals, such as HO_2 or RO_2 . Via a number of different atmospheric reactions some nitrogen oxides will finally become $\text{HNO}_3/\text{NO}_3^-$, which, with NO_2 are removed from the atmosphere via wet and dry deposition processes. Although it is not the major nitrogen oxide species in all areas, NO_2 is one of the most important air pollutants in urban areas, as it is the most significant nitrogen oxide species from a human health point of view.

Nitrogen oxides play a key role in the formation of photochemical oxidants, and this position paper will deal mainly with nitrogen dioxide and to a lesser extent, NO in air. The various sources, concentrations in urban and rural air, and the environmental effects of nitrogen oxides are considered. The environmental effects include human health effects, material damage and ecosystem (acidification and eutrophication) effects caused by NO_2 in the air, and deposition of nitrogen compounds. Effects on the ecosystems will mainly be the same, whether oxidised or reduced nitrogen compounds are deposited. For this reason, reduced nitrogen compounds will impact the assessment of oxidised nitrogen in the environment. A further factor is the interaction between nitrogen oxides and ammonia in the atmosphere.

1.2 Emissions of nitrogen oxides

Emission sources

European emissions of nitrogen oxides arise mainly from anthropogenic combustion sources, where NO is formed by reaction between the nitrogen and the oxygen in the combustion air and to some extent by oxidation of nitrogen in the fuel. The amounts formed depend on the pressure and temperature of the combustion process. Minor amounts of NO₂ (usually <10%) are emitted directly during combustion. Natural nitrogen oxides emissions emitted from soil, volcanoes and lightning are important on the global scale, but account for a small part (<10%) of total European emissions (Whelpdale, 1987).

The main anthropogenic sources are mobile combustion sources (road, air and water traffic) and stationary combustion sources (including industrial combustion). These main categories accounted for 57 and 39% respectively of the total European nitrogen oxide emission in 1994 (CORINAIR, 1995). Nitrogen oxides from industrial processes may be mainly emitted as NO₂, while the industrial combustion sources mainly emit NO. Directly emitted NO₂ may make significant contributions to the NO₂ concentrations in some areas.

Since the amount of NO_x emission is dependent on combustion conditions and process loads, estimates of emissions from different types of sources and areas will always be uncertain. In some cases the degree of uncertainty may be large.

Emission data and trends

Emission data from European States are inventoried and reported by the CORE Inventory AIR project (CORINAIR). Data for the year 1995 from major source categories in the different countries are presented in Table 1.1.

Official data on national nitrogen oxides emissions for Europe are reported to and compiled by European Monitoring and Evaluation Programme (EMEP) (Barrett et al., 1995, Barrett and Berge, 1996), set up under the UNECE Convention for Long Range Transboundary Air Pollution. Data have been reported since 1980 and the changes over time between 1980 and 1993 have been studied. The relative changes in emissions are presented in Table 1.2. In contrast to the trend for sulphur dioxide emissions, there has been a small reduction of 2% in total European nitrogen oxides emissions as a total for Europe.

Table 1.1 Emissions in the EU countries for 1995 by different source categories (CORINAIR, provisional data, July 1996). Units 1000 tons of NO₂ per year.

Source category	1	Combustion in energy and transformation industries
	2	Non-industrial combustion plants
	3	Combustion in manufacturing industry
	4	Production processes
	5	Extraction and distribution of fossil fuels / geothermal energy
	6	Solvent and other product industry
	7	Road transport
	8	Other mobile sources and machinery
	9	Waste treatment and disposal
	10	Agriculture and forestry, land use and wood stock change
	11	Nature

Emission source	Energy	Processes	Fuel, solvents	Mobile	other anthropogenic	natural	Total
Categories	1,2,3	4	5,6	7,8	9,10	11	
Country							
Austria	49	12	0	99	0	0	160
Belgium	148	4	0	218	4	0	374
Denmark	146	1	4	151	0	0	302
Finland	90	20	0	178	0	0	288
France	304	29	0	1132	29	6	1500
Germany	904	23	0	1455	0	0	2382
Greece	133	36	0	206	5	1	381
Iceland	1	0	0	22	0	0	22
Ireland	63	0	0	54	0	0	117
Italy	751	11	0	1365	31	0	2157
Liechtenstein	0	0	0	0	0	0	0
Luxembourg	11	0	0	10	0	0	21
Netherlands	176	12	1	317	27	10	543
Norway	44	8	0	170	0	0	222
Portugal	82	5	0	166	0	2	254
Spain	443	10	0	753	17	4	1228
Sweden	-	-	-	-	-	-	392
Switzerland	27	0	0	104	9	3	143
United Kingdom	852	5	109	1245	4	0	2216
Total	4224	177	114	7643	126	26	12310
%	34	1	1	62	1	0	100

Table 1.2 Emissions of nitrogen oxides in European countries in 1992 per area unit g NO₂/m²/year and within brackets in N/m²/year. The emission per inhabitant is given in kg NO₂ /person and within brackets as kg N per person. The data are based on official emission data reported by the countries to EMEP (Barrett et al. 1995). In addition, relatively large amounts of NO_x are emitted from ships on international water. No trend is estimated for these emissions.

Country	Emission per area unit	Emission per person	Relative change in total emissions (%) 1980 - 1993
Albania	0.3 (0.1)	3 (1)	0
Austria	2.6 (0.8)	30 (9)	-26
Belarus	1.3 (0.4)	26 (8)	-12
Belgium	10.8 (3.3)	33 (10)	-21
Bulgaria	2.3 (0.7)	30 (9)	-43
former Czechoslovakia	6.9 (2.1)	56 (17)	-33
Denmark	6.6 (2.0)	56 (17)	-4
Estonia	1.0 (0.3)	30 (9)	0
Finland	1.0 (0.3)	69 (21)	-4
France	3.3 (1.0)	33 (10)	-17
Germany	12.8 (3.9)	43 (13)	-17
Greece	5.6 (1.7)	76 (23)	0
Hungary	2.6 (0.8)	23 (7)	-33
Iceland	<0.2 (<0.05)	49 (15)	-8
Ireland	2.0 (0.6)	36 (11)	+67
Italy	5.9 (1.8)	30 (9)	+39
Latvia	0.7 (0.2)	16 (5)	0
Lithuania	1.0 (0.3)	20 (6)	0
Luxembourg	7.2 (2.2)	53 (16)	-17
Moldova	1.6 (0.5)	13 (4)	0
Netherlands	16.1 (4.9)	36 (11)	-4
Norway	0.7 (0.2)	56 (17)	+21
Poland	3.9 (1.2)	33 (10)	-24
Portugal	1.3 (0.4)	13 (4)	+48
Rumania	1.6 (0.5)	16 (5)	+20
Russia	<0.2 (<0.05)	20 (6)	+31
Spain	1.6 (0.5)	23 (7)	+32
Sweden	1.0 (0.3)	49 (15)	-6
Switzerland	4.3 (1.3)	26 (8)	-23
Turkey	0.3 (0.1)	3 (1)	0
Ukraine	1.6 (0.5)	20 (6)	-4
United Kingdom	11.2 (3.4)	49 (15)	-2
former Yugoslavia	1.6 (0.5)	16 (5)	+15

NO_x emissions from stationary sources have decreased in many areas, while the emissions from vehicles have increased due to the fact that growth in number of vehicles and in the distance travelled have been much larger than the decrease in emission factors (WHO, 1995C). Catalytic cleaning of car exhausts is a major improvement. It is, however, limited to new vehicles, and due to the relatively long life time of vehicles, it will take some time until "full effect" is achieved. Traffic sourced NO_x emissions are predicted to decrease from 1995 onwards under the influence of new vehicle technology (EC Air Quality Report of the Auto Oil Programme, DG XI 1996) and it is assumed that, within many Member States, NO_x emissions from stationary combustion sources are also decreasing and by 1998 will be reduced by 40% compared to 1980 emissions as a consequence of the Large Combustion Plants Directive 88/609. This decrease will be achieved by a number of measures including the introduction of low NO_x combustion technology, flue gas cleaning, fuel switching and nuclear energy.

Emission density

Emission data are reported country by country. Emissions over Europe is uneven. This is shown in figure 1.1 for 150 x 150 km grids. In urban areas traffic will generally make larger contributions to total NO_x emissions than at national levels. This is not reflected in such a large grid. For example in the UK national emissions from road traffic contribute about 50% of total emissions compared with over 85% in the West Midlands conurbation. At the street level the traffic contribution may be even greater. Table 1.2. shows emission data per person, per unit area and change in total emissions over the period 1980 to 1993. The spatial emission distribution for NO_x is also shown in Figure 1.1.

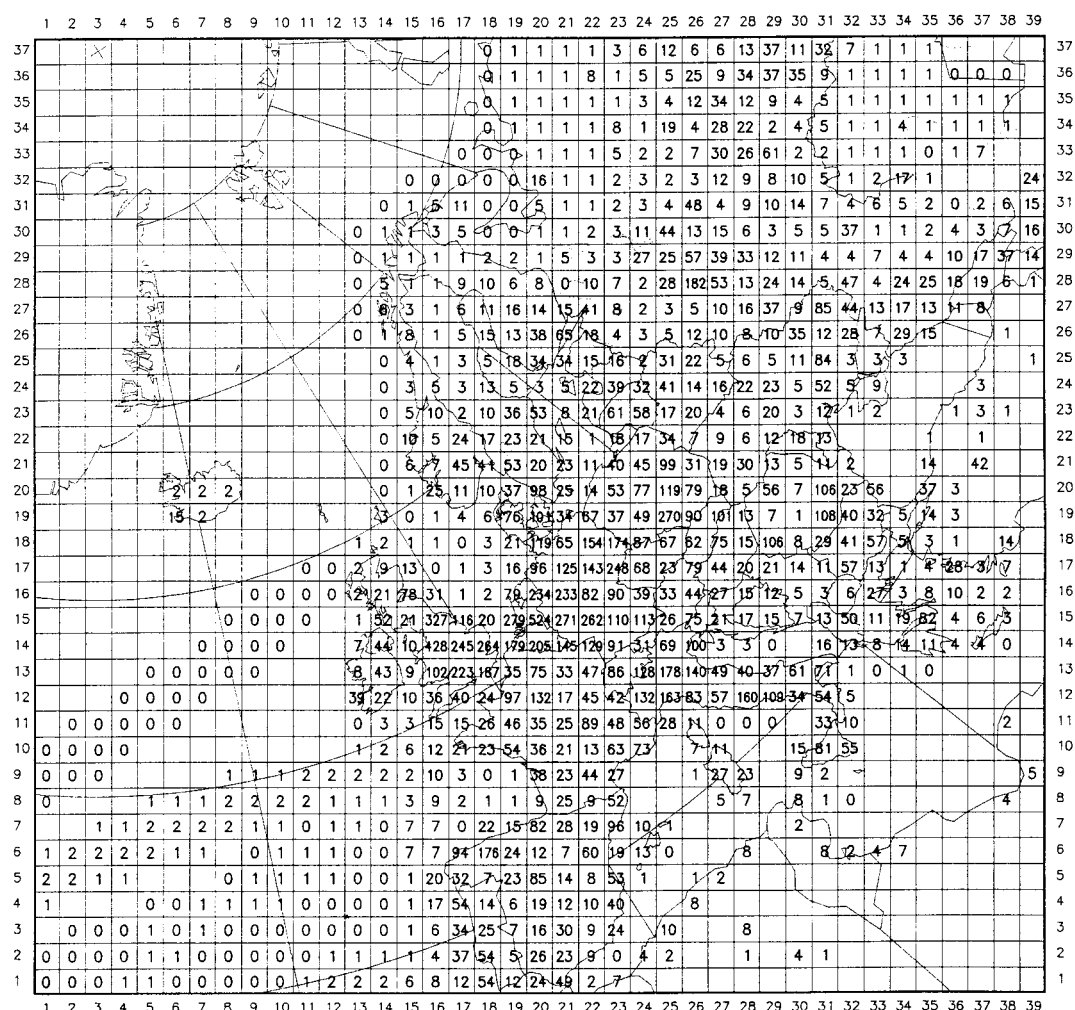


Figure 1.1 Emissions distributed spatially within the EMEP grid net over Europe in 1993. 1000 tonnes of NO_2 . (Barrett et al. 1995)

Nitrogen oxides protocol

Nitrogen deposition and its impacts on the ecosystems are dealt with by the UNECE Task Force on Mapping. The outcome of this work will form the main basis for the new nitrogen protocol. Abatement strategies as discussed for the new NO_x protocol, should take into account all aspects of air pollution. A complete assessment has not yet been carried out of which pollutants and which effects will require the most far-reaching reductions in different European regions in order to protect health, ecosystems and materials. The new NO_x protocol will mainly address the impacts of O_3 formation and deposition of nitrogen compounds, while the FWD will mainly address direct human health air exposure. The Nitrogen Oxides Protocol, under the UNECE Convention of Long-range Transboundary Air Pollution, is signed by 26 countries. It commits signatories to ensure that 1994 NO_x emissions do not exceed those in 1987. In addition, a number of countries committed themselves, to reduce their emissions by 30%. However, the majority of these countries are unlikely to achieve their targets. As can be seen in Table 1.2, few countries have reduced emissions more than 20%. A new nitrogen protocol is under preparation and negotiations are expected to begin during 1997. All effects of NO_x including human

health will be considered. Since many environmental effects, caused by nitrogen compounds are also caused by other pollutants, these other pollutants will also be considered producing a multi-pollutant, multi-effect protocol.

1.3 Atmospheric chemistry

Atmospheric transformation of nitrogen oxides

The atmospheric chemistry of oxidised nitrogen compounds is complicated, since they take part in a large number of chemical reactions and may be converted into a number of different chemical compounds. NO emissions are oxidised in the atmosphere to NO₂ and further to nitric acid/nitrate. NO₂ is present in urban air at concentrations between a few tenths to several hundreds of Tg/m³. Concentrations in rural areas are generally much lower and vary with distance from source. The concentration of NO₂ is a complex function of the meteorological situation, the emission rate of nitrogen oxides, the oxidation of NO to NO₂ and the oxidation rate of NO₂ to other species. Dispersion, deposition and chemical transformation cause the concentration of nitrogen oxides to decrease with distance from an emission source.

The cycle of nitrogen compounds is shown in figure 1.2.

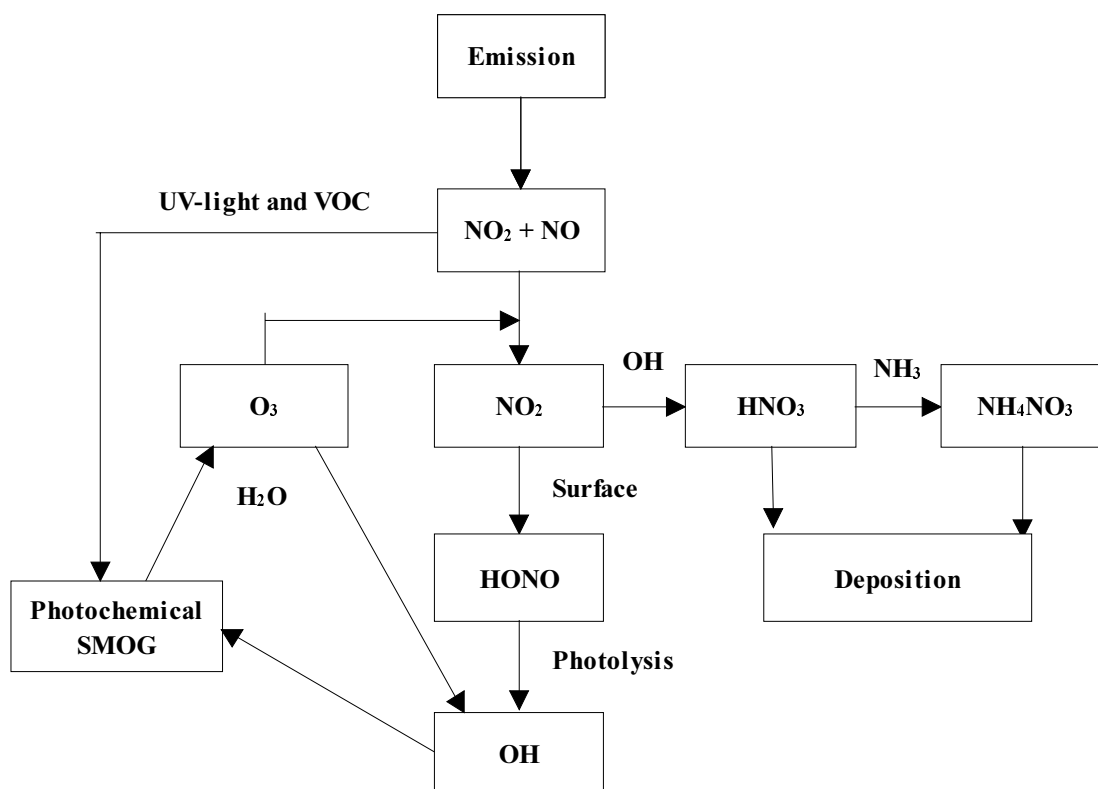
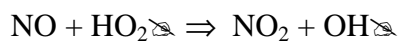
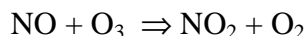
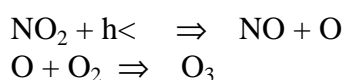


Figure 1.2 Simplified cycle of the nitrogen compounds in an urban environment

When released into the atmosphere, NO is rapidly oxidised to NO₂ through processes involving ozone (O₃) and, in highly polluted areas radicals, such as peroxy-radicals (RO₂) and the hydroperoxy radical (HO₂) according to the following reactions:

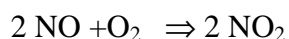


Oxidation by radicals is very fast and may cause high concentrations of nitrogen dioxide at roadsides (Allegrini & Febo, 1995), especially if high concentrations of HNO_2 provide OH radicals to trigger the oxidation process. In the presence of sunlight, NO_2 is photolytically decomposed back to NO with the subsequent formation of O_3 :



This leads to the photo-stationary state where the rate of oxidation of NO is balanced by its reformation by the photolytic dissociation of NO_2 . This balance is disturbed by the presence of VOC and other compounds which may be oxidised to peroxy radicals, and net ozone production may occur. At night the oxidation of NO by O_3 proceeds to completion, i.e. until either NO or O_3 is totally depleted.

NO can also be oxidised by molecular oxygen:



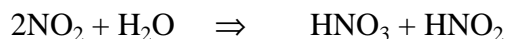
but this reaction is normally too slow to make significant contributions to ambient NO_2 levels. However, the reaction has a negative temperature coefficient and during persistent winter-time episodes at low temperature, when NO_2 levels may rise to $>650 \mu\text{g}\cdot\text{m}^{-3}$ (300 ppb) over several days, this process may lead to considerable contributions (Bower et al, 1994, Grennfelt, P. 1996).

NO_2 is removed from the atmosphere by several processes, mainly dry deposition and the oxidation to HNO_3 by OH radicals:

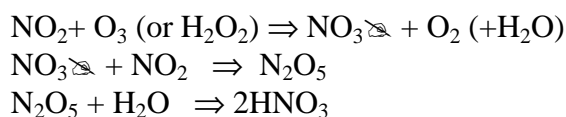


HNO_3 deposits rapidly on surfaces and also reacts easily with NH_3 giving ammonium nitrate (NH_4NO_3) in the particulate phase. HNO_3 is removed through dry and wet deposition processes.

NO_2 is also removed by heterogeneous reaction with water occurring on surfaces, generating HNO_2 and HNO_3 .



Photolysis only occurs during day. At night heterogeneous reaction between water and N_2O_5 produced via the NO_3 radical, is more important:



Particulate nitrate

Particulate nitrate, NO_3 , is formed by reaction of gaseous HNO_3 with sea salt, alkaline particles, or gaseous NH_3 or by reaction in liquid phase (e.g. droplets) of HNO_3 with alkaline components including ammonium and subsequent water evaporation. Conversion of gaseous HNO_3 and NH_3 to particulate NH_4NO_3 is a process of importance for deposition, since easily deposited gaseous compounds are turned into fine particulate of NO_3 and NH_4 , which are slowly deposited and transported over large distances. NH_4NO_3 and ammonium sulphate ($[\text{NH}_4]_2\text{SO}_4$) are hygroscopic, and humidity in the air will to some extent determine the size of the particles in the air and thus influence the rate of deposition. The formation of NH_4NO_3 is reversible and at low NH_3 concentrations can be converted back to HNO_3 . The supply of NH_3 is therefore an important factor in determining the residence time of nitrogen oxides.

Relation between NO_2 and ozone in urban air

The maximum NO_2 concentration in the lower atmosphere is limited by the presence of oxidising agent, mainly ozone to no more than about 100 Tg/m^3 (50 ppb) for most of the year although summer-time “ozone episodes” can lead to higher concentrations if they coincide with high NO_x concentrations. Direct emission of NO_2 , e.g. from tailpipe, can also lead to higher NO_2 concentration.

Ozone episodes occur when sunlight initiates a complicated photochemically driven chain of reactions in air polluted with VOCs and nitrogen oxides. The hydrocarbons are attacked by hydroxyl (OH) free radicals to form organic peroxy (RO_2) radicals which oxidise NO to NO_2 ; this, in turn, is photochemically decomposed back to NO with the subsequent formation of O_3 . Photochemical smog which can build up, often some distance downwind of the source area, may also contain increased concentrations of hydrogen peroxide (H_2O_2), peroxyacetyl-nitrate (PAN), other hydrocarbon reaction products and particulates.

Air Pollution Climatology

Climate and, in particular, ventilation (air flow and mixing height) are as important in determining atmospheric concentrations as are emission rates. Thus the characteristics of the structure of the wind fields and the dynamic evolution of the boundary layer are key elements in the understanding of urban atmospheric pollution.

In urban areas the ventilation processes cannot be described because complex terrain and thermal disturbance modify them. For example it can result in the formation of heat islands, which often form around urban areas, and can hinder an efficient exchange of air with the surroundings. In such conditions, air pollutants are re-circulated in the boundary layer and concentrations can become high. In the re-circulated air masses, photochemical reactions may occur and worsen the situation. This is especially the case in the Mediterranean area, where land-sea breezes causing recirculation, are both common and strong.

The dispersion capacity of the atmosphere has been described from measurements of ambient short-lived radioactivity levels associated with the decay of radon (Allegrini

et al., 1994). The use of this technique in several urban areas has shown that the occurrence of high concentrations of nitrogen dioxide may be associated with radical reactions, taking place in the lower boundary layer during stable atmospheric conditions. When the atmosphere is unstable, most NO₂ is formed through the titration of NO with O₃. In that case, the maximum expected concentration of NO₂ is given by the O₃ concentration in the free troposphere, the layer above the boundary layer (Febo et al., 1996).

Conclusion

In conclusion, the atmospheric chemistry of nitrogen compounds is complex and in urban areas, is also dependent on the presence of other pollutants, such as hydrocarbons, and on meteorology. Effects will also differ with differing climate. There is no linear relationship between emissions of NO_x and concentrations of NO₂. As a result, it is difficult to quantify the emission reduction needed to reduce NO₂ concentration to any given level.

1.4 Concentrations in ambient air

Nitrogen dioxide in urban air

Urban concentrations of nitrogen oxides are frequently higher than in other areas and near emission sources. There is often a steep gradient in NO₂ concentration from the roadside to the more general urban background. This gradient may be less pronounced in heavily polluted areas and during episodes.

There are four recent large studies reviewing the air pollutant concentrations in European cities: WHO study "Concern for Europe's Tomorrow" (CET), RIVM/NILU Study on Air Quality in Europe (the "Dobris Assessment") (Sluyter, ed., 1995), the Auto-Oil study in 7 cities by the EC and the European Topic Centre on Air Quality, MA2-4 project report "European Air Quality, 1993" (Larssen et al., 1995) .

In the WHO (CET study), a calculation of the urban population exposure to NO₂ is presented. NO₂ concentrations from 144 cities in 18 European countries are available for the late 1980s. The cities represent 91 million people, 29% of the European urban population. The annual average concentrations of NO₂ exceeded 60 Tg/m³ for 19% of the residents in these cities and 100 Tg/m³ in one city with about one million inhabitants.

In 25 of the cities reported by the WHO CET study, inhabited by 21 million people, daily average NO₂ concentration exceeded the WHO AQG (1987) of 150 Tg/m³. In most cities the exceedances occurred less than 10 times per year and were more frequent in Western than in Eastern European cities.

Urban air concentrations of NO₂ in major European cities 1985 - 1990, are summarised in Sluyter, ed (1995). Only cities with more than 500,000 inhabitants were included in contrast to the WHO study which included smaller cities. Annual average NO₂ concentrations were similar in many of the Western European cities. During 1990 the mean of the annual NO₂ concentrations in 22 cities was 46 Tg/m³

with a standard deviation of only 6 Tg/m³. The average concentrations in 26 cities in former Soviet Union were the same, 46 Tg/m³, but the variations were larger; with a standard deviation of 22 Tg/m³.

Sluyter, ed., (1995) reported that exceedances of the 1987 (24 h) WHO AQG of 150 Tg/m³ for NO₂ were observed at background locations in 12 of 40 cities from which data on daily measurements throughout the year were available. The maximum 24 hour average concentration at a city background site reported from these 40 cities in 1989-91 was 616 Tg/m³. The maximum 1 hour average concentration was 532 Tg/m³. The maximum 24-hour value and the maximum 1-hour value from the 40 cities, were reported from different monitoring sites.

The report by the European Topic Centre on Air Quality, on European Air Quality 1993 (Larssen et al., 1995) contained NO₂ data from 90 cities (194 sites in total, of which 80 are in Germany). Most of the cities can be classified as urban background sites, but traffic exposed sites are probably also represented in the data. Annex VI gives an overview of the measured concentrations for the 50-percentile (median), 98-percentile and maximum 1 hour.

The extent of exceedances in 1993 of the present EU guide values and WHO Air Quality Guidelines are given in annex V. For practical reasons, only a selection of the data reported to European Environment Agency (EEA) is presented in the tables. The highest concentrations reported were 98 Tg/m³ for the 50-percentile, 253 Tg/m³ for 24 hour average and 554 Tg/m³ for 1 hour average, all reported in Athens. There were exceedances at the following number of measurement sites, table 1.3.

Table 1.3 Number of exceedances in the ETC/AQ study (Larssen et al., 1995).

		Number of sites in exceedance	Total number of monitoring stations
EU Guide values	Tg/m ³		
50-percentile (median)	50	34	163
98-percentile	135	13	163
WHO Guidelines	Tg/m ³		
max. 24 hour average	150	5	109
max. 1 hour average	400	3	97

These data show that in the general urban atmosphere of cities in Europe, the median guideline, is exceeded most frequently, with the highest value 95 Tg/m³. 98-percentile and 24 hour and 1 hour guidelines are exceeded less often

However, hot spots, i.e. street sites, are not well represented in the data base. Therefore, these data do not give a representative picture of the extent of exceedances of short-term (1 hour) average concentrations in Europe.

Traffic is the main contributor in Western European cities and space/domestic heating in small boiler units the dominating source for NO₂ in the former Soviet Union. The

emissions in Northern European cities seem to be lower than in Western and Eastern Europe. The Southern and Central European cities seem to have slightly higher annual average concentrations. However, these differences have not been shown to be statistically significant.

The data indicate that at least 21 million people in Europe are exposed to daily NO₂ concentrations above the WHO AQG (1987) level, 150 µg·m⁻³. If these data represent the air quality in cities where no measurements are made, 20% of the European urban population are exposed to levels exceeding the WHO AQG (1987). However, parts of the population may be exposed to higher levels of indoor NO₂ pollution. In 1996 WHO adopted new air quality guidelines, including a one-hour guideline of 200 µg/m³ and an annual guideline of 40 µg/m³ for NO₂.

The Auto-Oil study was carried out in Athens, Cologne, London, Lyons, Madrid, Milan and The Hague. Data available from 1980 onwards showed that one or more EU or WHO air quality objectives have been exceeded on occasion in all of the cities studied. Air quality modelling carried out as part of the work indicates the emission reduction necessary to comply with the new WHO-AQG for NO₂ in some European cities. Between 20 and 50% NO_x emission reduction is necessary; the highest figure relates to cities in southern Europe (Auto Oil Report, 1996).

Concentrations near traffic

The highest concentrations in urban areas are alongside busy roads. Measurements show that roadside NO₂ levels can exceed WHO AQGs (1987) by a factor of 2 - 4 depending on actual traffic and dispersion conditions. Persons living close to or working in the street environment are often highly exposed to nitrogen oxides and other traffic-related pollutants.

Concentrations in rural background areas.

Model calculations using the EMEP model (Barrett et al., 1995) indicate that high NO₂ concentration is mainly an urban pollution problem. The concentration of NO₂ in rural areas does not exceed the 1987 daily WHO AQG (150 µg/m³), except in areas, where local emission sources influence the situation (Figure 1.3).

Trends in NO₂ concentrations

As with the data on emissions, no obvious general trends in air concentrations of nitrogen oxides in most European cities have been observed. Small decreases in NO₂ concentrations are indicated in Austria and Switzerland, where NO_x emissions were reduced by almost 30%. Decreases were also indicated at Swedish urban background sites after correction of monitoring data for the milder climatic conditions during recent years (Sjödin et al., 1995). Small decreases were observed at some urban areas in Denmark. Generally, it has not been possible to collect comparable data from many countries to estimate the trends.

Secondary NO_x compounds in the air

Secondary NO_x compounds, and other oxidants formed by oxidation of NO, may also have a considerable environmental impact, which has to be considered in NO_x abatement strategies. In many European ecosystems, effects of nitrogen deposition and photochemical oxidants are of greater importance than the effects of high concentrations of NO₂.

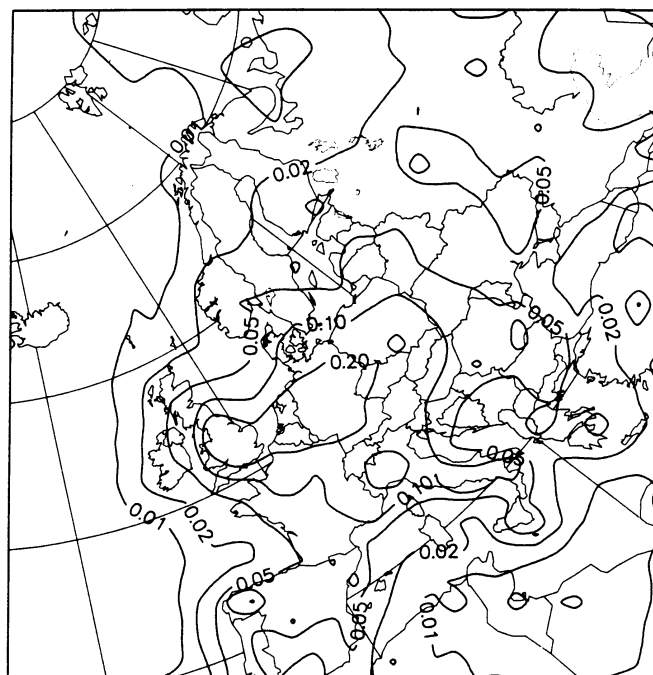
Deposition of nitrogen compounds contributes to acidification and eutrophication of sensitive ecosystems. The deposition of nitrogen over Europe calculated by the EMEP model (Barrett and Berge, 1996) show a maximum deposition near the major source areas. Deposition of nitrogen oxides is from the ecosystem effect point of view added to the deposition of reduced nitrogen.

The deposition of nitrogen and the photochemical oxidants is not only a local but also a transboundary air pollution problem. In order to decrease the transboundary pollution effects caused by NO_x emission, a freezing of the emissions was adopted within UNECE, i.e. the NO_x protocol to the Convention on Long Range Transport Air Pollution. The setting of new limits for urban air quality of NO₂ will lead to emission reductions which are also necessary for protection of the ecosystems from nitrogen deposition exceeding critical loads.

Indoor concentrations of NO₂

In the absence of indoor sources the indoor concentrations are 30 to 100% of the outdoor concentrations. For NO₂, additional indoor contributions may occur from gas fuelled stoves, ovens and water heaters and smoking. These contributions may be considerable in relation to the outdoor pollution and must be considered from a total health assessment point of view. As mentioned in the "scope" section of this document, indoor pollution is not the object of this position paper.

Mean Concentration of Nitrogen Monoxide

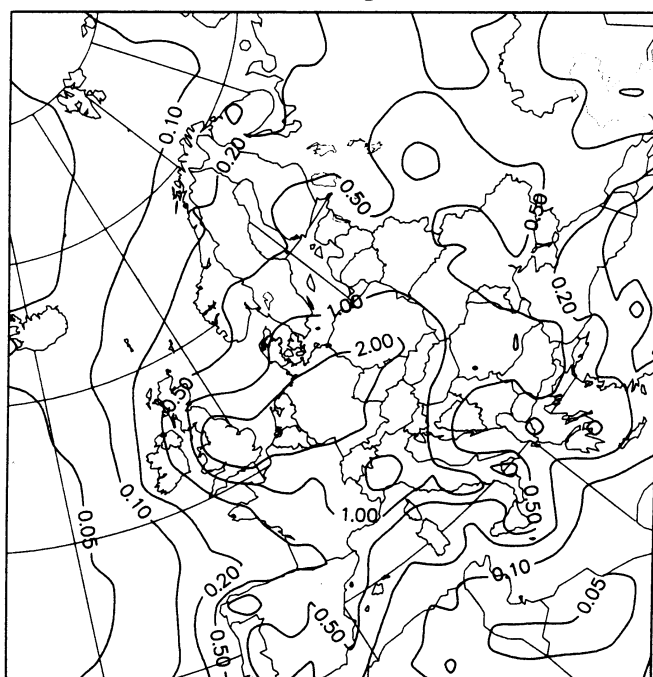


Year: 1994

Isolines:

0.01
0.02
0.05
0.1
0.2
0.5

Mean Concentration of Nitrogen Dioxide



Year: 1994

Isolines:

0.05
0.1
0.2
0.5
1.
2.
5.
ug(N)/m3

Figure 1.3. Mean concentration of NO_2 over Europe 1994 (Barrett & Berge 1996). The concentrations are given as $\mu\text{g N/m}^3$. To transform it to $\mu\text{g NO}_2/\text{m}^3$, multiply by 3.3.

2. Risk assessment

2.1 Impact of nitrogen dioxide

The Directive on Ambient Air Quality Assessment and Management requires the establishment of limit values and/or alert thresholds. When setting the limit values and, if appropriate, an alert threshold, the following factors may be taken into account:

- the degree of exposure of sectors of the population, and in particular sensitive groups;
- climatic conditions;
- sensitivity of flora and fauna and other habitats;
- historic heritage exposed to pollutants;
- economic and technical feasibility;
- long-range transport of pollutants, and secondary pollutants, including ozone.

Consideration must be given to the total impact of nitrogen oxides emissions on human health, materials and ecosystems from different nitrogen oxide compounds and other secondary pollutants, e.g. O₃.

Nitrogen deposition and its impacts on the ecosystems are dealt with by the UNECE Task Force on Mapping. The outcome of this work will form one of the main bases for the new nitrogen protocol. Abatement strategies as discussed for the new NO_x protocol, should take into account all aspects of air pollution. A complete assessment has not yet been carried out of which pollutants and which effects will require the most far-reaching reductions in different European regions in order to protect health, ecosystems and materials. The new NO_x protocol will mainly be based on the impacts of O₃ formation and deposition of nitrogen compounds. FWD will mainly take care of the direct air exposure.

2.2 Human health

Short and long term exposure to NO₂ can induce health effects. The health effects related to the two types of exposure are often different in character. Short term exposure to a very high NO₂ concentration can result in severe pulmonary damage in healthy humans. Exposures of persons with chronic lung diseases such as asthma and chronic obstructive pulmonary disease (COPD) can cause short term responses such as changes in lung function or airway responsiveness. Chronic (i.e. long term) exposure to NO₂ has been associated with increased respiratory symptoms, especially when observed under indoor exposure. The methodologies are also generally quite different for the study of acute reversible conditions and chronic health effects. Thus, it is pertinent to separate effects occurring after short term and long term exposure in the evaluation (Berglund et al. 1993). However, it should be realised that it is sometimes difficult to determine whether observed health effects result from short or long term exposure.

Some studies have shown that synergistic effects of NO₂ and O₃ may exist (Kagawa and Tsuru, 1978). NO₂ and O₃ in combination are suspected to exacerbate the allergic reactions to inhaled allergens. Similar responses are shown in animal studies (see for example Abraham et al. 1983, Matsura 1970 a,b,c. and Yanai et al. 1990). In the recently presented Europe-wide study on "Short Term Effects of Air Pollution on Health: a European approach using epidemiological time series data" - The APHEA project, (St Leger, 1996, ed.) epidemiological effects, particularly mortality and hospital admission were analysed from the air pollution perspective. Relations between pollution levels and health effects were observed, but the role of NO₂ was not clear.

In the 1980s WHO reviewed the literature on the effects of air pollution on human health and recommended guidelines. The first set of guidelines were published in 1987. (WHO Regional Publications, European Series No. 23, 1987). WHO adopted new Air Quality Guidelines in October 1996. They will be published shortly. The following sections discuss the work leading to these revised guidelines and recent work on eco-toxic effects by the UNECE.

WHO and UNECE recommendations

WHO has concluded that despite the large number of acute human exposure studies there is no evidence for a clearly defined concentration/response relationship for NO₂ exposure. For acute exposures, only very high concentrations (> 1880 Tg/m³) effect healthy people. Asthmatics and patients with chronic obstructive pulmonary diseases are clearly more susceptible to acute changes in lung functions, increased airway responsiveness, and symptoms. Based on small changes in lung functions and changes in airway responsiveness in several studies, a range of 375 to 565 Tg/m³ over a 1- to 2-hour exposure is a clear lowest-observed-adverse-effect level (LOAEL). A 50 % margin of uncertainty is included in the WHO recommendations because of the reported statistically significant increase in response to increased airway responsiveness with exposure to 190 Tg/m³ and a meta analysis suggesting changes in airway responsiveness. Based on human clinical data, a 1-hour guideline of 200 Tg/m³ is recommended as a maximum value. At double this recommended guideline, there is evidence to suggest possible small effects in pulmonary function of asthmatics. Should an asthmatic be exposed either simultaneously or sequentially to NO₂ and an aero-allergen, the risk of an exacerbated response to the allergen is increased. At 50 % of the suggested guideline (i.e. 100 Tg/m³) no acute exposure studies have been undertaken, so effects are unknown.

WHO has recommended an annual average guideline of 40 Tg/m³ (WHO, 1996) Although there is no particular study or set of studies that support a numerical value, the WHO's database clearly indicates the need to protect the public from chronic exposures. For example, indoor air studies with strong NO₂ sources (e.g. gas stoves) suggest that an increment of about 30 Tg/m³ (two week average) is associated with a 20 % increase in lower respiratory illness of children 5 to 12 years old. However, the effected children had a pattern of indoor exposure that included peak exposures higher than those typically encountered outdoors. Thus, they cannot be extrapolated quantitatively to outdoor air pollution. Outdoor epidemiological studies suggest a range of potential effects, providing support for the indoor studies, although they do

not provide consistent exposure information for NO₂. Furthermore, animal toxicological studies show that prolonged exposures can cause decreases in lung host defences and changes in lung structure. Based on expert judgement 40 Tg/m³ is proposed as an annual average guideline.

WHO (WHO A+B, 1995) has recommended the following guidelines for NO₂, i.e. 200 Tg/m³ as 1-hour average and 40 Tg/m³ as annual average. WHO has not considered recommendations for alert thresholds.

Exposure above WHO-AQG.

The EEA's Topic Centre for Air Quality under has collected air quality data for Europe (Air Quality in Major European Cities, R.J.C.F. Sluyter (Ed.), 1995). The report includes estimates and classification of population exposure and exceedances of WHO-AQGs for SO₂, O₃ and PM, but not for NO₂. A very rough estimate of the NO₂ level has been made on the basis of data from Dutch cities. Based on this estimates of exposure above WHO-AQGs have been made.

WHO has also made exposure estimates for NO₂, (WHO C 1995). The estimates are made on the basis of outdoor concentrations, and it was assumed that these were representative for the air pollution levels people are exposed during their daily activities. The outdoor levels were estimated from measurements and air quality modelling.

The estimated percentage of the population exposed to concentrations above the WHO 1987 24 hour AQG in the studied cities is shown in table 2.1. No estimates are available for exposure above new (1996) WHO AQG.

2.3 Effects on vegetation by nitrogen oxides

Nitrogen oxides are absorbed by vegetation in the same way as CO₂ through stomata. Nitrogen oxides are dissolved in the stomata cavity water and form nitrite and nitrate, which in turn are reduced to NH₃ and eventually incorporated into organic compounds. (e. g. Wellburn, A.R., Wilson, J., Aldrige, P.H. 1980). If too much NO₂ is absorbed over time, acute damage may occur in form of necrosis. Biological membranes (e. g. Mudd et al. 1984) and chloroplasts (Wellburn et al. 1972, Lopata & Ulrich, 1975) are assumed to be damaged. Acute effects occur at very high concentrations, which are seldom observed in ambient air, except near very large point sources (Stonybrook Lab., 1994). There is a range of long term exposure effects (Guderian and Tingey, 1987). Up to a certain level, no effects are observed. Above this, NO₂ may stimulate growth. However, higher doses will decrease growth in relation to controls. There is at present a dispute over which nitrogen oxides are the most toxic. Further knowledge is necessary to assess the situation.

Table 2.1 Exposure of the populations of cities with data to annual mean concentrations of NO₂ (upper table). Exposure of the populations of cities with data to daily NO₂ concentrations above 150 µg/m³. 1986-1991 (lower table)*

Annual mean NO ₂ concentration (µg/m ³)	Percentage of city populations exposure							
	Western countries		CCEE		USSR		All countries	
	Up to 1985	After 1985	Up to 1985	After 1985	Up to 1985	After 1985	Up to 1985	After 1985
< 60*	79.7	76.1	61.5	75.9	80.8	96.3	79.2	80.9
60-100	20.3	23.9	0.0	15.4	12.0	3.7	16.2	17.9
> 100	0.0	0.0	38.5	8.7	7.2	0.0	4.6	1.3
Number of people in cities with data (millions)	(32.8)	(56.2)	(3.0)	(13.2)	(20.1)	(21.7)	(55.9)	(91.1)

* No WHO air quality guideline: 60 Tg/m³ is the lowest level at which long-term exposure is reported to cause respiratory effects.

Countries	Exposure (average number of days per year)	Percentage of the population exposed for at least 1 day	Number of people in cities with data (millions)
Western countries	13.4	27.9	56.2
CCEE	32.4	24.1	13.2
USSR	5.4	9.7	21.7
All countries	15.4	23.1	91.1

* WHO air quality guideline (24-hour mean concentration)

The effect nitrogen oxides on vegetation depends on the action of NO and NO₂. The ratio of NO₂ to NO is very important for the deposition rate, since NO₂ is deposited much faster than NO. Indirectly, NO may also have a positive effect on vegetation by consuming O₃. The NO₂ has a lower phyto-toxic effect than O₃.

Recommendations made by UNECE and WHO to protect vegetation

Critical levels for oxides of nitrogen in the atmosphere and critical loads for nitrogen deposition have been developed within UN ECE (Caporn, 1992) to protect ecosystems. Guidelines to protect ecosystems have also been adopted by WHO, based on the UNECE work. It is pointed out that synergistic effects (or multi pollutant effects) are very important and the guideline is given for NO_x rather than NO₂ alone. WHO recommended a guideline to protect vegetation as an annual average of 30 Tg/m³ for NO_x, calculated as the sum of NO and NO₂ in ppb and expressed as NO₂ in µg/m³. Existing guidelines and limit values for protection of vegetation and sensitive ecosystems are summarised in Annex 2.

2.4 Effects on ecosystems by oxidised nitrogen compounds

Any change in nitrogen deposited to an ecosystem will cause a change in the nutrient status of the system. It will cause biological consequences such as favouring nitrogen-loving species, eutrophication and in the most severe case, acidification. The acidification is, in turn, linked to leakage of nitrates to ground and surface waters, as the system cannot consume all the nitrogen deposited. Indirectly, O₃ formed via NO_x emissions may also damage vegetation, crops, natural vegetation and forests.

The development of recommendations for limit values based on critical loads is outside the scope of this paper. Limit values for NO₂ and NO_x will however have implications for acidification and O₃. These should be taken into account in assessing the potential impact of the recommendations in this position paper. As mentioned above, nitrogen deposition and its effects on the ecosystems are dealt with within the UN ECE Task Force on Mapping and the mapping work lead by Co-ordinating Centre for Effects at RIVM in the Netherlands. The outcome of this work will form one of the main bases for the new nitrogen protocol. Effects of ozone are dealt with by another working group.

2.5 Effects on materials

The effect on materials of an individual air pollutant is difficult to separate from the effect of others. Many of the effect on materials have been ascribed to the multi-pollutant situation in the urban atmosphere than to the action of separate pollutants. Nitrogen oxides may contribute to damage in several ways, but their role is not yet clarified. However, the role of nitrogen oxides is secondary compared to that of ozone for dyes, fabrics, plastics and elastomers, to that of oxygen, sulphur dioxide and wetting by rain, condensation, mist etc. on metals and to that of rain, carbon dioxide and sulphur dioxide for calcareous stonework. There is also a possibility that nitrate deposited on outdoor surfaces, can enhance the microbial activity and thus increase bio-degradation. Increased growth of algae due to deposition of nitrogen compounds on surface is considered as a soiling and undesirable effect. Nitrogen containing compounds have also been recognised as a possible problem in museums (De Santis & Allegrini, 1992).

Within the UNECE Integrated Co-operative Programme on Materials a large number of exposure sites have been set up, where pollution concentrations and material deterioration rates are measured. The programme aims to quantify the effect of SO₂ in combination with NO₂ and other pollutants. So far no results are available that clarify the role of NO₂. The main reason being SO₂ and NO₂ tend to be both present in urban air. However, the ratio of nitrogen to sulphur oxides is increasing at many sites, and, this may mean nitrogen oxides may exert a more substantial effect on material damage in the future.

At present there is not sufficient data to enable a recommendation to be made for an air quality limit value for NO₂ in order to protect material from air pollution damage.

2.6 Recommendations by the working group

Directive 96/62/EC requires that limit values and when relevant alert thresholds should be established with the aim of avoiding, preventing or reducing harmful effects on human health and the environment as a whole. Where possible therefore such objectives should be based on the Lowest-Observed-Adverse-Effect-Levels (LOAEL) on human health and the ecosystems, taking sensitive subgroups/ ecosystems into account. The Directive does not provide for non-mandatory guide values or target values.

The Directive recognises that the actual air quality at many locations in the Members States may not comply with the new limit values or alert values, but based on technical and economical considerations a time of compliance will be proposed in accordance with the analysis made by a separate study on cost implications.. The principles are illustrated in figure 2.1.

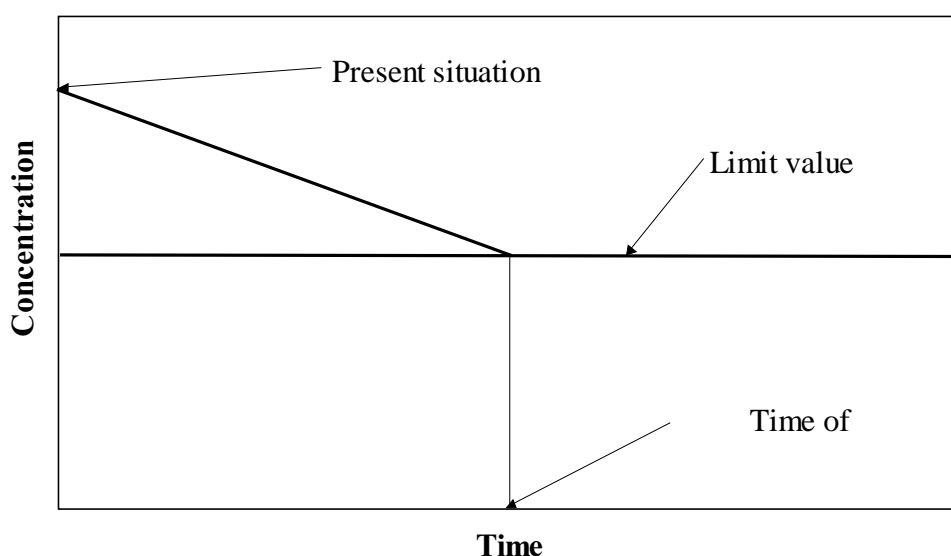


Figure 2.1 An illustration of the principles of the establishment of limit values based only on effects and acceptable/possible time of compliance based on economical and technical possibilities.

Recommended air quality limit values

The proposal for the new air quality limit values (table 2.2) was based on the recommendations WHO health assessment, i.e. the update and revision of the Air Quality Guidelines for Europe (WHO, 1987 and WHO, 1995A) and the ecological assessment from the meeting in les Diablerets (WHO, 1995B).

Table 2.2 Air quality limit values for NO₂ proposed by the majority of the working group (exclusive industry); the measurement period is one year.

Type of value	Averaging time	Statistical parameter	Level	Pollutant
Short term limit value	1 hour	Not to be exceeded more than 8 hours per year *)	200 µg/m ³	NO ₂
Long term limit value	annual	average	40 µg/m ³	NO ₂
Limit values to protect vegetation	annual	average	30 µg/m ³	NO _x =NO+NO ₂ **))

*) 8 hours per year is approximately equal to 99.9-percentile. The value cannot be directly compared to the present limit value which is 200 µg/m³ defined as the 98-percentile.

**) Added as ppb and expressed as NO₂ in µg/m³.

WHO has defined its short term limit guideline a maximum value, while the present EU limit value is a 98-percentile. The maximum value is very sensitive to measurement errors, but the 98-percentile is difficult for the public to understand. The majority of the working group has recommended to define the short term limit value as a value, which can be exceeded 8 times per year (99.9-percentile). In the discussions in the Working Group and in the Steering Group, the question of introducing a lower concentration with compliance at the 98- percentile has been discussed several times. With respect to air quality the 99.9-percentile on 200 µg/m³ corresponds to a 98-percentile on 90-120 µg/m³, somewhat varying between areas depending on emissions and other pollution characteristics. The Auto Oil report (1996) estimates the corresponding 98-percentile 93 µg/m³. At the same time the Working Group recommend, that the member states should apply additional tools for quality control and the assessment of exceedances in addition to measurement data, e.g. statistical methods, emission inventories and air quality models, comparisons between different stations etc. These topics will be treated in chapter 3 "Measurements and assessment of concentrations" of this position paper. The exceedances of the air quality standards must be assessed by the Member States and reported to the Commission. Not only exceedances should be reported, but also statistical parameters of the concentration frequency distribution, e.g. percentiles, averages etc., i.e. for comparison with the present limit values.

The long term limit values are recommended as annual averages.

The Working Group realised that the measured concentration of the air pollutants including NO₂ depends very much on the location of the monitoring sites. It was emphasised that data from different types of sites; high altitude sites, urban background, residential and rural background areas, are relevant for estimates of the human exposure, for assessment of the contribution from different sources and for understanding of the chemical and physical processes, controlling air quality. The

Working Group has considered siting criteria in relation to the above limit values and alert thresholds based on lowest detectable effects (chapter 3).

The Working Group agreed to give examples of concentrations, which may serve as alert or information thresholds (Table 2.3). These should be directly health related and increase the awareness of the general public. It was concluded that it will be confusing to define an information threshold that can apply generally all over Europe from south to north and from large urban areas to smaller residential areas, because the air quality is different. The Working Group considered defining alert thresholds as well as information thresholds using the principles of the O₃ directive. Table 2.3 gives examples of thresholds and the corresponding observed effects of exceedings. Several Member States have set different alert values. Denmark has set an alert values of 350 Tg/m³ for 1 hour average, but the alert is only issued, if the episode is expected to last more than 12 hours. Austria has set a pre-alert value of 350 Tg/m³ and also alert levels at 600 and 800 Tg/m³ as 3-hour means.

Table 2.3 Examples of alert levels chosen with consideration only to their effect from a health point of view.

Threshold	Effect considered
200 Tg/m ³	According to a Swedish study this level is the lowest effect level for asthmatics (Bylin, 1993)
400 Tg/m ³	Based on the WHO assessment that levels of 375 - 565 Tg/m ³ give small but statistically significant reversible effects on lung function and airway responsiveness in mild asthmatics.

The Working Group did not however, recommend an alert thresholds for NO₂. . This was because it was felt that an alert threshold value should be the same as the limit value as the latter is based upon lowest observed effect level. Also short term measures to reduce concentration, for example by restricting traffic, are unlikely to be effective because NO₂ is mainly a secondary pollutant. The Working Group therefore recommended the provision of better information to the public, when the short term limit value is exceeded, see chapter 3.6.

3. Measurement and assessment of concentrations

3.1 Introduction

When limit values (and alert thresholds) are set, the FWD states, that ambient air quality shall be assessed in each Member State. In accordance with FWD, measurement is mandatory in zones with a population of more than 250.000 inhabitants, with high population concentration (agglomerations) and where the concentrations are near to or exceed the limit value, see table 2.4. and figure 3.1.

Table 2.4. Summary of requirements for measurements and other assessment methods

Type of Zone	Assessment technique
Agglomeration, >250.000 inhabitants	Monitoring is mandatory even if levels are <x% of limit value
Zones with levels >x% of limit value	Monitoring is mandatory
Zones with levels <x% but >y% of limit value	A combination of measurements and modelling may be used.
Zones with levels <y% of limit value	Modelling or objective estimation is sufficient.

Determination of X and Y

A consistent method of deriving the X (higher assessment level) and Y (lower assessment level) is requested from the Commission. The working groups for the different daughter directives have suggested X and Y to be based on the inter-annual variations:

$$(1 + 2*SD)*X*LV \leq LV$$

which gives

$$X = \frac{1}{(1 + 2 * SD)}$$

where SD is the normalised standard deviation of data at monitoring stations. Based on the data in the APIS database it is estimated that there is a slight variation between stations, but the standard deviation is generally consistent for NO₂ throughout the European Union.

For Y the criteria used is 3 times standard deviation.

which gives

$$Y = \frac{1}{(1 + 3 * SD)}$$

For NO₂ the data in the Air Pollution Information Service (APIS database have been used to calculate X and Y, Table 2.5 The span given is the approximate range calculated. The recommended value is given without brackets.

Table 2.5 Estimates of X and Y based on data from the APIS database

	X	Y
annual value	(70) - 80%	(65) - 75%
1 hour values	60 - (70)%	50 - (60)%

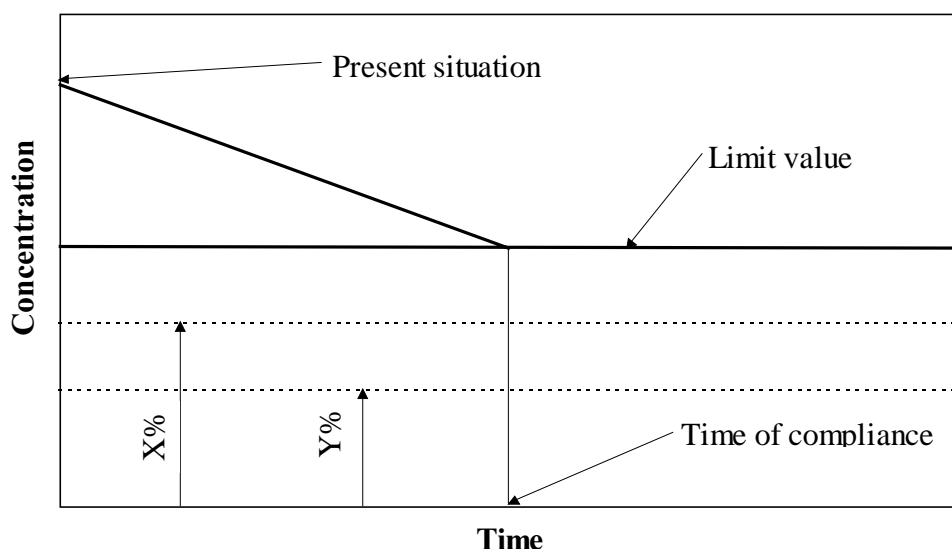


Figure 3.1. The use of X% and Y% values. Measurements shall be made in zones, where the pollution level is expected to be higher than X% of the limit value. If the level is lower than Y% of the limit value, model or objective estimation can be used. Between X% and Y% of the limit value a combination of measurements, campaigns and modelling can be used for assessment of the air pollution.

If the concentrations are low, a combination of measurements and models may be used in non-agglomerations. A combined use of measurements and models may also reduce the monitoring need in agglomerations .

The FWD also states the requirements for the assessment of air quality, such as design of monitoring programmes, requirements of the methods to be used etc.

There are several reasons for assessing nitrogen oxides in the air, the most important being to identify the levels of NO₂ in areas where people are likely to be exposed, and to identify areas where air quality standards are exceeded. For the abatement of pollution, the identification of major sources is necessary, as well as monitoring the effectiveness of applied abatement strategies.

Due to the close relationship between NO and NO₂, both should however be measured, to provide a basis for abatement strategies. It is also useful to measure O₃ in connection with the measurements of nitrogen oxides due to the "titration" of NO. Recommendations for monitoring O₃ will be included in a future position paper for O₃.

To evaluate the large scale concentrations of nitrogen oxides, it is useful to measure at least NO₂ and O₃ in rural background air. Such measurements are made in most countries in Europe within the EMEP programme. In many areas in Europe, urban monitoring networks to be supported by rural background sites to separate rural background concentrations from local urban pollution for abatement planning.

For monitoring in urban areas, it is necessary to distinguish between urban background and roadside. "Roadside" sites are those close to the traffic sources. "Urban background" are sites at some distance from streets, mainly influenced by the total pollution load in the urban air rather than by a few specific sources. Distinction between the two types of sites is of importance for the estimation of population exposure. Most people spend the majority of their time outside exposed to air pollution concentrations more like urban background than those found at the roadside. Additional measurements may be needed in residential areas which are not characterised as urban background, but may be subject also to the influence of traffic and industrial pollution.

Many people are exposed for short periods to high concentrations of NO₂ when travelling by car, bus, bicycle or on foot. Smaller groups of the population may spend considerable time in the street environment and thus be heavily exposed to traffic exhaust, include. NO₂. The difference between roadside and urban background is also of great importance for the selection of control measures. Decreasing the general NO₂ in urban background will require emission reductions, whereas improvement of air quality at roadside may be obtained merely by changing traffic pattern.

3.2 Description of monitoring methods

The monitoring of NO₂ for regulatory purposes requires methods suitable measuring concentrations in the range relevant for Europe, i.e. 0 - 500 µg/m³, with a sufficiently high accuracy. There are several well-established monitoring methods for NO₂ in this concentration range, some of which can only measure NO₂, while others can also measure NO and/or NO_x. A list of methods for NO₂ measurements has been prepared by JRC and is presented in Annex III. The list includes references to the methods as well as brief comments on their advantages and disadvantages. The methods differ with respect to chemical and technical principle as well as to temporal and spatial resolution. In some situations, only one monitoring method may be feasible, but where a choice of method is possible, the advantages and disadvantages of different methods need to be considered.

The most commonly used methods are briefly described below. Distinction is made between continuous and discontinuous methods, and between point measurements and remote sensing. The point measurements are representative for the monitoring site and for an area around the site, the size being determined by the emissions and other characteristics of the surroundings. The remote sensing methods measure the average concentration over the monitoring path in the atmosphere. Variation along the path is not detected.

Many other methods are available. However, only standardised and reliable methods should be used for environmental monitoring.

Discontinuous point measurements

Active and passive discontinuous methods are normally used for daily to monthly sampling. The manual discontinuous methods are relatively cheap and simple, but have several disadvantages including the need for manpower for sampling and analysis, the limited time resolution and the time delay until results are available.

For the daily measurements of NO₂, two active methods, the potassium iodide and the Saltzmann methods have. Both methods can be operated on a 24-hour basis, although for the Saltzmann-method, the sampling time is recommended to be shorter. These daily sampling are used in many European rural areas, for example in the EMEP programme. In some countries it is also the main monitoring method at urban background sites.

The potassium iodide method is based on the absorption of NO₂ on a potassium iodide impregnated sintered glass filters (Ferm and Sjödin 1993). NO₂ is absorbed and reduced to nitrite by the iodide on the filter. The nitrite formed is extracted with de-ionised water and determined spectro-photometrically with the Griess method.

The Saltzmann/modified Saltzmann method is based on the direct Griess reaction during sampling (ISO, 1981). A pink colour is produced during sampling. The intensity is measured spectro-photometrically (Mücke et al., 1995).

More details are found, e.g. in the new EMEP manual (EMEP-CCC1995) and in the GEMS/AIR's - WMO, UNEP handbooks.

Diffusion denuders combined with an analysis by ion chromatography may also be used for the determination of NO₂. Carbon coated denuders allow for the simultaneous determination of NO₂ and PAN at concentration levels which approach 50 ppt (parts per trillion) on a 24 hour basis which makes the technique very suitable for the measurement of NO₂ in rural areas (De Santis et al., 1996).

Diffusive, or passive, sampling methods are normally used for longer sampling periods, on weekly (usually in more polluted areas) or monthly (usually in less polluted areas) basis. The sampling technique is based on molecular diffusion of the gas molecules into the sampler, where they are quantitatively collected on an impregnated filter. No electricity, pump or other equipment (except for a rain shelter) is needed. The method is used routinely for NO₂ (Svanberg et al., (1994) and (1995); Atkins et al., (1995); Campbell et al., (1994), Brauer and Brook, (1995).

Diffusive sampling is suitable for two or three-dimensional mapping - weekly to monthly measurements - of air concentrations in an urban area. Diffusive samplers are simple and cheap and the procedure can produce a large amount of long term air quality data to a fairly low cost. In combination with other measurements with higher

temporal resolution, the passive sampling is valuable for the total air quality assessment. However the diffusive samplers need validation.

The NO₂ collected on the impregnated filter is extracted from the filter with water and determined by chemical analysis. When calculating the concentration, the ambient air temperature must be taken into consideration, since the diffusion velocity is temperature dependent. Studies on the accuracy of diffusive samplers in comparison with other monitoring techniques for NO₂ show a good agreement with chemiluminescence measurements, as well as with the potassium iodide method (Svanberg et al., 1994, Ferm & Svanberg 1997), (Saeger, 1996). The diffusive sampling technique for other urban pollutants, such as NO has also been recently developed by the Ogawa Co, and by other organisations (Ferm and Sjödin, 1993; Saeger, 1996), for SO₂ (Carmichael et al. 1995; Ferm and Svanberg, 1997), for VOC (Svanberg et al., 1994, 1995) and for O₃ (Koutrakis et al., 1993).

Continuous point measurements

Continuous monitoring methods cover various instrumental techniques. However, the chemiluminescence method is the recommended and most commonly used method. The instrument signals are recorded continuously intervals and typically integrated to hourly means.

Chemiluminescence. The technique is based on the gas phase chemiluminescence reaction of NO with O₃, which produces stimulated NO₂ emitting light (chemiluminescence) at about 1200 nm wavelength (ISO, 1985). The different types of chemiluminescence monitors measure NO_x and NO concentrations on the air, using a somewhat different design. The air sample passes a heated catalytic converter where NO₂ is reduced to NO. The signal from the air passing the converter gives the NO_x concentration, while the signal from the air drawn directly to the detector gives the NO concentration. NO₂ is determined as the difference between the two signals. The instrument is commonly used in urban air and is the reference method recommended by United States Environmental Protection Agency, US EPA, 1991).

The sensitivity of the traditional commercial monitors is usually not sufficient for rural, low-polluted areas. The method is also unspecific. Other reducible nitrogen compounds (such as HNO₃ and PAN) may interfere. This is of less importance in urban air, where concentrations of NO and NO₂ dominate.

Remote sensing, DOAS

DOAS (Differential Optical Absorption Spectroscopy) is an open path optical measuring technique applicable for a number of gases, including NO₂ specifically, absorbing light in the UV and visible spectral regions (Platt and Perner, 1983). The technique is based on differential absorption, i.e. the difference between local maximums and minima in the absorption spectrum of the probed gas. The DOAS technique may also be used for NO, but a shorter path is required than for NO₂.

Light from a broad-band xenon high-pressure lamp is transmitted up to several kilometres through the atmosphere. The light is received and analysed by the use of a fast scanning dispersive spectrometer to eliminate the influence of air turbulence. The DOAS technique is used today in a large number of applications, the most common

being environmental monitoring of the main pollutants in urban areas, as a mean concentration over some distance in urban air. The method has recently been approved by US EPA for SO₂, NO₂ and O₃.

The major advantage of DOAS is that several compounds can be measured at the same time, i.e. HNO₂, NO₃ radicals, benzene, toluene etc. The DOAS technique is not yet US EPA approved for these pollutants, but by application of suitable quality control it will give reliable results. However, the scattering of light by fog, heavy rain or snowfall may cause problems with data quality and availability. For aromatics significant interference must be taken into account.

The light absorption over the entire light path is integrated, which gives an average concentration over this path. This makes the technique less suitable for measurements very close to sources (e.g. in a street) and not directly comparable with point measurements at such sites. However, the technique is suitable for measurement of urban background concentration, where the small scale spatial variations from small local sources are not relevant or directly describing the information about the urban background air pollution. The calibration of the instrument is easy and stable over long time periods.

Recommendations for reference method

Reference methods for ambient air monitoring should fulfil the following criteria:

- be accurate and precise
- fully comply with the Directive requirements
- give data in the correct time scale
- be easily applied and operated

The chemiluminescence method is recommended as reference method for mandatory concentration measurements. It can easily validate other methods. Equivalent methods may be used, if they are shown to be in agreement with the reference method.

The reference method must be calibrated correctly, see chapter 3.5 to produce good quality results.

3.3 Network design and siting criteria

The aims and objectives of the network required by the EU are clearly stated in the FWD. The legal requirements of the Directive for assessment of ambient air quality are given in Article 5. In addition, the aims of the Directive, as given in Article 1, also need to be considered:

- establish objectives for ambient air quality in the EU designed to limit or prevent harmful effects to the environment as a whole and to human health
- assess the ambient air quality in Member States in a uniform manner
- make available to the public information on ambient air quality
- maintain good ambient air quality and improve poor ambient air quality

In order to "assess the ambient air quality in Member States in a uniform manner" the network should be similarly designed in all Members States. These sites could then be regarded as the European Ambient Air Quality Directive Network (EAAQDN) analogous to the EMEP monitoring network for rural areas in Europe.

The information on air quality should be made available to the public and decision makers. The data will be reported to the Commission in accordance with Article 11 (2). In addition the number of exceedances of alert values will be advised.

The national or local requirements for the network may be different, but to minimise the burden on the Member States, already established networks may, as far as possible be accommodated within the requirements of the Directive. New networks should be established in accordance with a harmonised network design and operation.

The aims of the network

During the design of monitoring networks account needs to be taken of both the number of sites and their location. It must reflect the need to measure exposure of humans and the environment. All site groups in Table 3.1 should be considered for inclusion in the network.

Since automatic monitoring is very expensive, it will sometimes be preferable to use automatic monitoring in combination with indicative monitoring (passive sampling) and/or air quality modelling to more cost effective assess the extent of high concentration areas and the spatial variability of pollutants.

Table 3.1 The site groupings for NO₂.

urban traffic	urban/residential	industrial	rural background
street (roadside)	central city	boundary fence	suburban
street canyon	suburban	residential	rural
tunnels	residential	max. impact	remote
central city	commercial		agriculture
suburban			
residential			

Since the limit values for exposure of humans will be for 1 hour and annual concentrations, the measurements and other assessment tools should be applied for these averaging times. The highest peak values of NO₂ occur in street canyons, where traffic is the main source and it is most likely that the short term value will be exceeded here. The residence time is generally short in street canyons, but many people may be present, when the concentration is high. The long term exposure may be more critical at residential or suburban areas.

The indoor air quality is outside the scope of the FWD, but the indoor air quality is significantly influenced by the outdoor air quality, which means that outdoor air quality monitoring is relevant for estimates of the contribution to the indoor exposure.

NO₂ concentrations at different sites

The suggested design of the network is i.e. based on experiences with design of networks in Italy (Allegrini, 1996), Denmark (Palmgren, 1995) and discussions in the working group. The NO₂ levels in an urban area is a result of,

- the rural background air pollution, which is the NO₂ pollution transported over long distances, e.g. from other urban areas or by recirculation in the region. The air pollutants have had time to react with other components of the atmosphere. The rural background concentration is measured away from local emission sources.
- the urban background air pollution, which is the rural background plus the air pollution from the urban area. This air pollution is a result of the emissions in the city as a whole, the dispersion conditions and many chemical reactions. ,
- the local air pollution, which is the air pollution close to the emission sources, e.g. traffic or point sources . Only the fastest chemical reactions are important for the local emitted pollutants. The concentrations of these pollutants are mainly determined by the dispersion.

Air pollution at all three levels must be taken into account in order to obtain a total description of the NO₂ pollution in an urban area.

The rural background can be difficult to estimate, depending on the climatic conditions. In northern Europe the dispersion of pollutants is mainly determined by the air flow due to the prevailing wind and surface roughness. In the southern Europe the dispersion is highly influenced by thermal convection and local wind flow systems. On the large scale, especially in the southern part of Europe, the dispersion is more complicated, due to complex flow structure. The long residence time may also lead to more complicated chemical processes, which are not easy to describe (see chapter 1.3). In these cases the models for dispersion and chemical reactions need further development and present knowledge is mostly based on measurements.

The urban background can be measured by few measurement stations located away from local sources, e.g. above the roof tops, in traffic free squares, in parks etc. The stations should represent the air pollution level, without significant impact from a single source (e.g. a local point source or a street) The representativity of such stations for the whole urban area must be documented, for example by using passive samplers to determine optimal locations. The optimal location is defined as the location that represents the average concentration of a certain area.

The local air pollution depends very much on the local sources, e.g. the traffic in the street or point sources and the local dispersion conditions, and micro-scale influence on the aerodynamics of the site e.g. by the surrounding buildings.

The main properties of the wind flow in an urban area and especially in a street canyon are well known. When the wind direction is perpendicular to the street direction, a vortex is generated in the street canyon, whereby the wind flow at street level is opposite to the flow above the roof level. Such a wind circulation results in a characteristic dependence of pollution in the street on wind direction. Concentrations on the windward side are much lower than those on the leeward side.

The simple and very important fastest chemical processes in the local scale involving NO, NO₂ and O₃ are well known. The residence time of the air in the street canyon can be estimated and from this the transformation to NO₂ of the emitted NO from the traffic in the street.

However, many slower processes are insufficiently understood to allow their current use in model simulations, e.g. the formation of radicals and the transformation of VOCs in a large urban area or a region.

The total description of local air quality by measurements requires many monitoring stations. However, many suitable models exist for the calculation of air pollution dispersion in local scale, and for fast chemical reactions, providing all relevant urban background air quality data and local meteorological parameters are available (*Berkowicz et al., 1994*). Consequently, it is not necessary to measure at all highly polluted streets. Only measurements at typical streets are necessary and the results transferred to other streets by model estimates. These tools are very important for air quality planning.

Definition of site types in and around urban areas.

Based on the above considerations it is recommended that the monitoring network for urban sites should be planned to include five types of stations:

- Rural background sites for evaluation of pollution levels in rural and remote areas caused by near-by large sources and/or remote sources, regional air pollution including photochemical air pollution.
- Urban background, where all controlled pollutants are measured.
- Urban residential area sites, where pollutants of relevance for protection of health are measured.
- Local Industrial sites near large industrial installations
- Local traffic sites, street sites, for evaluation of pollutants emitted directly from vehicles.

The air pollution at rural/remote sites should be measured. The regional air pollution can be measured at stations outside greater urban areas. It is in many countries measured within the EMEP programme.

The urban background station is important, not only for producing information on exceedances in the urban area. It can also measure other pollutants, and can produce data relevant for the understanding of the physical and chemical processes occurring in urban air and for following air quality trends. For understanding of the processes it is especially necessary to measure O₃ at the sites.

The residential sites, which are important for estimation of exposure of the population, often coincide with the urban background sites, but may also be influenced significantly by point sources or traffic.

The air pollution around industrial sites is regulated in many countries by local authorities. It is not possible to determine the air quality around point sources and possible exceedances of short term limit values are not possible to determine without a large number of monitoring stations. The best way is to combine emission estimates and dispersion modelling with measurements at a few sites.

In the street environment, measurements are performed near the building line and no more than 5 meters from the carriageway. The pollution at street sites is defined by the traffic characteristics (traffic density and composition, average speed etc.), the buildings or other obstacles along the street and the orientation of the street. Sites along highways through unbuilt and non sensitive areas are not relevant. Measurements should be carried out at selected typical zones and a total description of the exposure, e.g. of inhabitants should be carried out by model calculations.

For management purpose it is recommended that O_3 and NO should be measured in addition to NO_2 at both rural and urban background sites in order to describe the NO/NO_x pollution.

Micro scale considerations for site locations

For selecting the detailed location of sampling sites, at the micro scale, a large number of additional factors also need to be considered:

- height of the inlet: at street sites not lower than 2-3 metres above ground; in urban background: inlet can be higher, if the site is representative for a larger area
- sheltering from trees, buildings etc.: well away from obstacles such as trees, buildings etc.; street sites near to the building line and no more than 5 metres from the carriageway
- distance from crossing, traffic lights etc.: more than 25 metres
- manifold design

In addition, the following should be considered:

- interfering sources
- security
- access
- electrical power and telephone
- "visibility" of the site (aesthetic)
- safety of public and operators
- planning requirements

Number of sites and other tools

The overall number of monitoring stations established will depend on:

- ambient concentrations, in relation to limit values
- the population likely to be exposed to such concentrations
- the scale and extent of environmental effects
- the spatial variability of NO₂

The number of stations will depend on the size of the urban area, but at least one of each type of station should be selected in zones where the ambient air quality must be assessed in accordance with FWD Art. 6. The Working Group has proposed the number of stations required based mainly on experience from existing networks.

When no other assessments are applied, the number of stations recommended N_i can be determined as:

Urban background : $N_i = 2 + (I * 0.4)^{1/2}$

Local: $N_i = 1 + (I * 8)^{1/2}$;

where I is the number of inhabitants (in millions) in the agglomeration.

The minimum number of stations (N_0) can be chosen, if supplementary selected assessment (e.g. models, campaigns etc.) are used to increase information about the air quality.

Of these sites, at least one should be an urban background site, where NO₂, NO and O₃ concentrations are measured as one hour averages. It should be selected in the centre of the traffic zone. The monitors could be chemiluminescence or DOAS. DOAS is a good choice, if a long measurement path can be established, because it gives more representative data of the urban background. In larger urban areas more urban background sites of the above type should be selected, e.g. one site for every 50 km² of the urban "core". Outside the urban "core" at urban background sites and in suburban areas with lower source density the sampling can be carried out with much less time resolution, e.g. by diffusive samplers.

Between 3 and 10 street sites should be selected depending on the size of the urban area and the diversity of the street types, and generally close to the urban background stations. The busy streets (canyons) with the highest expected pollution with NO₂ pollution and where many people are exposed (living, working or transported) should be selected. At street sites the chemiluminescence method should be used for measurements of NO and NO₂. The measured data should be used for generalisations and NO₂ concentrations in other streets estimated by street pollution models.

Measurements around point sources should be carried out in accordance with the requirements laid down for licensing new plant in the Member States. It may include application of point source models.

Monitoring of the NO₂ concentrations in order to protect vegetation should be made in rural areas, outside the urban zones and agglomerations and in zones defined as "ecosystems".

The main elements of monitoring and assessment are summarised in table 3.2.

Table 3.2 The main strategy of monitoring and assessment and the number of stations (N_i) and minimum number (N_0) as it is recommended by the Working Group.

Type of station	N_i	N_0	Location	Comments
Urban background	2 to 4	2	Representative for large areas (e.g. in parks, at roof tops etc.)	Locations close to city centre and at least one residential areas should be selected, possibly partly supplemented with measurement campaigns
Local	1 to 10	1	Roadside at different typical street types	Supplemented by street pollution modelling at other locations and in other streets
Industrial	1 to ?	1	Determined by model estimates	Application of pollution modelling is necessary. A large number is necessary if only measurements are used
Rural	1 to n	1 to n_0	Determined by model estimates	The number, n, should be 1 station per 20000-50000 km ² depending on the density of sources. n_0 corresponds to 50000 km ²

For design and optimisation of the monitoring network it is recommended that the mandatory measurements are combined with indicative methods, continuously or in campaigns.

For monitoring long time average concentrations, discontinuous measurements, especially diffusive sampling is recommended since it is a cost-effective and practical method for air quality surveillance. It is also well suited for the surveillance of the spatial distribution of air pollution. (See also chapter 3.6).

3.4 Other assessment techniques

To support air quality assessment in the Member States, the Commission asked the EEA and the Joint Research Centre European Reference Laboratory on Air Pollution (JRC-ERLAP) to develop a Guidance Document (van Aalst et al. 1997) linked to the FWD.

The FWD defines zones as part of a Member State territory, to be defined by the Member State, and agglomerations as zones with a population of more than 250,000 inhabitants or with sufficiently high population density to justify separate air quality assessment and management. This Guidance Document provides criteria, procedures and methodology for four assessment cases:

1. Preliminary assessments

According to the FWD article 5, Member States, who do not have representative measurements of the level of pollutants for all zones and agglomerations, shall undertake series of representative measurements, surveys or assessments, in order to have data available in time for implementation of the daughter Directives.

2. Assessments supporting further station siting optimisation of (mandatory) measurements

For implementation of the daughter Directives, 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 reliably throughout a Member State.

3. Assessments supporting generalisation of (mandatory) measurements

In zones where concentrations show many potential areas of exceedance, for instance in busy streets in an agglomeration, stations at one location could represent similar locations elsewhere. This is referred to as generalisation.

4. Evaluating existing assessments

Member States may have existing data on air quality for some or all zones and agglomerations. The Guidance document should also provide criteria for Member States and the Commission to judge whether such existing information is sufficient for a preliminary assessment (FWD, Article 5), or whether additional work should be carried out.

There are three main assessment tools. Besides measurements emission inventories and model calculations are used to describe the air quality (van Aalst et al., 1997).

Assessment of air quality is an important issue in many areas, especially in the urban environment. The most credible, complete and useful assessment is obtained by combining measurements and modelling results. The monitoring at chosen sites provides data on air quality and its temporal variability. Dispersion models extend the knowledge by computing concentration fields as a function of time at various spatial scales. Under the Directive, if air quality is close to the limit value modelling

alone can not, and may not, be used to assess the air quality situation and exceedances of air quality standards.

Models are also useful for

- Exploratory assessments in absence of measurements and for planning of measurements and location of monitoring sites.
- Understanding the relationship between air quality and factors such as special dispersion conditions and temporal and spatial emission variability.
- Relating air pollution to sources and sectors as input information assisting the development of abatement strategies.
- Calculating pollution levels as a consequence of future emission scenarios.

A number of air pollution models have been developed for different spatial and temporal levels of urban concentrations, and for the various applications mentioned above. The models can be separated into four classes:

- models for air quality near streets and roads
- models for air quality near point sources
- integrated urban scale models, taking account of all source categories, and giving concentrations in a grid net and in selected receptor points.
- population exposure models

For the evaluation of possible air quality standards exceedances, the integrated urban models are the most useful. Single point source models will only provide information on the contribution from a point source, which is of limited use for a total assessment. Since traffic emissions will to a great extent determine the urban air quality, street pollution models are useful to assess the zones at risk for exceedances, and for the evaluation and extrapolation of measurement results to larger zones. This type of model is relatively simple and is easy to use. The results are, however, only indications of the real situations and must be evaluated further using monitoring data and more advanced models.

Although there are a number of well-established models in use in urban areas in different parts of Europe, no general recommendation can be made here. However, the model must fulfil the requirements of QA/QC (see van Aalst, 1997) and be thoroughly validated against monitoring data.

An increasing demand for air quality assessment is foreseen and will be the driving force towards further development and improvement of dispersion models. Various models are needed. Operational models are an important tool for city planning. On-line models are used for air pollution forecasting at pollution episodes to show present and near-future risks. Models for regulatory purposes are used to demonstrate zones of compliance/non-compliance with air pollution directives.

To be powerful tools, the models must fulfil certain quality criteria. Work on quality assurance of models is important and has recently been started within different organisations. Accuracy can be documented in terms of uncertainties in input quantities and in model formulation, and could be demonstrated in the validation

process. Reference models will be defined in time. Development, validation and application of urban models will be carried out within EUROTRAC-2, SATURN.

Input, in the form of emissions and dispersion conditions, is necessary at a similar spatial and temporal basis as the output concentrations to be calculated by the model. Data on emissions is required with sufficient spatial and temporal resolution to describe the important gradients occurring in a zone.

Emission inventories are part of the modelling activity. Methodology for making inventories have been developed within the EU CORINE work, EUROTRAC GENEMIS and UNECE EMEP. The harmonisation and standardisation of models within DGXI, DGXII, European Environment Agency - Topic Centre for Air Quality, at a number of international organisations and on national scale is progressing. A compilation is made in Annex VII. The Guidance Report (van Aalst et al. 1997) will provide valuable information and guidance on which models to use and how to apply them.

In the assessment of models, monitoring data serve as validation. The validity of the modelling results depends on the accuracy of the input data as well as on model performance.

3.5 Calibration and quality assurance

Data quality objectives

Minimum levels of time coverage and data capture, at the necessary levels of precision, must be established in order to comply with the monitoring objectives. Based on the results of the NO₂ Quality Assurance Programme, organised by the JRC from 1992 to 1995, the precision and accuracy (expressed as maximum uncertainty) can be estimated as approx. $\pm 25\%$. This figure takes into account various uncertainties, including sampling losses (5%), calibration errors - primary calibration, transfer standard, routine calibration under field conditions - ($\pm 15\%$), analytical errors - such as noise, non-linearity, effect of atmospheric pressure and temperature- (5%) and the effect of interfering gases - water vapour, other nitrogen species - ($\pm 10\%$).

However, it should be noted that QA/QC programmes are currently only implemented efficiently in a few monitoring networks, and the quality of the measurements can be significantly improved by adoption of requirements. The following data quality objectives that can be achieved through the implementation of this QA/QC programme are:

- Continuous or semi-continuous measurements: 15% (individual measurements)
- Indicative measurements: 25% (individual measurements)
- Modelling 30 - 50% (may be different for daily, monthly or yearly averages)
- Objective estimation: 50 to 100%

The time coverage for continuous measurements should be 100%, excluding calibration time. For indicative measurements a minimum of 50% should be covered

(every second day, or at random, or 6 months per year for example). A lower coverage can be discussed, but it must be ensured that the temporal distribution of samples yields a statistically sound long-term mean. Minimum data capture for continuous measurements are 90%. Breakdown of instrument , excluding calibrations, allowance is 36 days per year.

Further details on required QA/QC will be given in the Guidance Report (van Aalst et al. 1997).

The measurements of NO₂ can be divided into three separate steps; sampling, analysis/measurement and calibration (when the analysis method is not absolute). The tables in Annex III give the currently used methods and their main advantages and disadvantages.

Requirements are also specified for the network management structure and to fulfil the requirements with respect to

- national focal point
- national co-ordination of data
- harmonised methodology for statistical evaluation
- minimum data capture
- standardised reporting directive
- exchange of information decision

The calibration and quality assurance of the national networks should be based on common requirements in the Member States. The activities on these topics should be co-ordinated by ERLAP, JRC. Generic capability transfer and harmonisation must include operational manuals and guide books on site operation, QA/QC and data handling. Routines must be developed for training, workshops, information exchange and technology.

Quality assurance and quality control of measurements

Quality assurance is a system of procedures that ensures that measurements are precise and accurate. Further, results must be comparable and traceable, and data representative of ambient conditions. The QA/QC system will ensure that optimal use is made of resources.

The major constituents of a quality assurance programme concern network design (see chapter 3.3) are, the selection of measurement technique - sampling, analytical and calibration procedure-, selection of equipment and methods for evaluation validation of methods and the test of instrument performance. Routine site operation as well as calibration under field condition, maintenance, management and training must be developed and documented.

QA/QC procedures examples are described in WHO UNEP GEMS/AIR Methodology Review Handbook Series, Volume 1, "Quality Assurance in Urban Air Quality Monitoring" and in the EC "Instruction Manual for Air Pollution Monitoring: Nitrogen Dioxide Monitoring".

Currently, QA/QC programmes only exist in a few monitoring networks of the EU Member States and with a variable degree of efficiency, as demonstrated by the recent field intercomparison organised by ERLAP in 36 network stations.

The variable quality of the existing laboratories, as well as the increase in numbers expected as a consequence of the implementation of the FWD, and the need for consistency across the network suggests the establishment of a laboratory accreditation process.

Different 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 EN45001 procedure was developed by CEN in collaboration with the Commission and is best adopted for testing laboratories in the field of air pollution measurements. Laboratories asking for accreditation are audited by a national or international accreditation organisation. This audit mainly concerns aspects such as laboratory installations and equipment, qualification and training of personal, proper quality control, technical audit and traceability of the measurements. The requirement for laboratory accreditation is the only enforceable way to ensure an effective QA/QC procedure

As validation of the European Network measurement methods and instrument certification is essential, the development of standard CEN procedures is necessary.

The Commission must organise EU wide inter-laboratory comparison exercises (round robin tests, inter-laboratory exercises, spot checks in the monitoring networks) to ensure comparability of measurements at international level. The QA/QC of measurements must contain audits including control of the operation of monitoring equipment at the sites, maintenance and calibration in the laboratories and data control.

Operational control must be in accordance with the above manuals and guide books. It is especially useful for NO₂, to have data on O₃ and NO available, see chapter 1.3. A plot of NO₂ versus NO_x concentrations will detect elementary errors in the measurements. The sum of NO₂ and O₃ (defined as O_x) will be relatively constant in a larger area (Palmgren et al, 1995).

Quality assurance and quality control of models.

Harmonisation and QA/QC of the application of models are being carried out under projects managed by the European Topic Centre on Air Quality. The tasks are to collect and review information on the requirements of EEA for air pollution models and model applications and provide guidance for selection and consistent application of validated models. See further in the Van Aalst et al. (1997)

3.6 Data evaluation and presentation for assessing the exceedance of limit and alert values

The monitoring data from the network have to be transmitted in accordance with the FWD, Article 4 (1) and the data will be published in accordance with Article 4 (2). The Directive also states that air quality shall be evaluated for the total zone of the Member State in accordance with Article 6. The quality and spatial representativity of monitoring sites and data should be reported. Good documentation of monitoring procedures and sites are necessary.

The three different types of monitoring stations are representative for different zones. The rural background stations with long averaging time are representative for large zones and it is possible to report any exceedances for large areas, typically several hundreds of square kilometres.

The urban background stations are representative for parts of the urban zone. The zone of representativity can be estimated by model calculations or indicative measurements by diffusive sampling. The data shall be reported with a defined zone of expected exceedances.

The urban traffic stations in the street environment are representative for the site of the street, but the measured data can be generalised to the entire street by street pollution models. In other streets the concentrations of NO₂ can be estimated by street models based on traffic data, meteorological data and the street geometry. The data should if possible be generalised and could be reported for the entire urban zone as km of street with exceedances. It is not possible in this working group to work out in detail the models to be used for this purpose, but it is recommended to establish a small expert group in co-operation with the Topic Centre, for preparation of a detailed operational street pollution model based on the existing street models.

In addition, for comparison with earlier measurements, the data shall be reported in accordance with the old NO₂-directive, i.e. the 98-percentile and the median of one hour averages for all monitoring stations.

Information on air quality to the public

All information on air quality shall be available to the public and mass media. As means to involve the public in the efforts to fulfil the aims of the FWD, responsible authorities shall actively and regularly disseminate information on the air quality. Appropriate ways can include e.g. summarising papers but also day-by-day reports in newspapers, local radio and television or continuous screens presentations at public locations etc. To improve the understanding and concern for air quality the development of simple methods for school children, citizen groups etc. to study and monitor the situation shall be encouraged (Nilsson, 1995).

Air quality management planning

The first stage of air quality management is a complete view of existing air quality. The Member States should evaluate and interpret the data, to understand the connection between sources and pollution patterns in time and space. Models - for single sources, for example point sources and traffic exhausts - and multi-source models - for large areas, urban or other - as well as indicative measurements are valuable tools in this process. The zone of exceedance is a key quantity to be assessed.

The models to be used for assessment of the air quality can also be used for planning, i.e. calculations on different emission scenarios. These could be used for selection of different measures to be taken for improvement of the air quality and for assessments of the effects of already taken actions. In addition, they can be used for prognoses for the air quality based on meteorological forecasts.

Traffic exhaust models can be applied, not only for NO_x and NO₂ in the street environment, but also for other traffic related pollutants, i.e. CO and benzene.

4. Cost implications

4.1 Introduction

To support the work of the Working Groups and the Commission, an "Economic Evaluation of Air Quality Targets for Sulphur Dioxide, Nitrogen Dioxide, Fine and Suspended Particulate Matter and Lead" was initiated. The study was not finished before this position paper was prepared, and so its results and conclusions have not been taken into account in this chapter. The chapter is therefore based on the material collected by the Working Group, and on the knowledge and experience of the Group Members.

Evidence in chapter 2 suggest that current concentrations exceed the proposed limit values in different areas in Europe, and it is necessary to assess the effect of planned measures to determine what abatement strategies may be required to meet the limit values. Comparison of the present air pollution situation with an of the impact of current (and planned) legislation will enable the development of the new strategy. Consideration of different Member States (chapter 1.4 and Annex IV) current pollution levels and exceedances of recommended limit values indicates that further reductions in NO₂ levels of approximately 30%, are needed in many large urban areas. From these data it is only possible to give an uncertain estimate on the need for NO_x emission reductions. A better estimate can only be made by more detailed model calculations taking dispersion, deposition and atmospheric chemistry into consideration.

The first NO_x protocol will end in 1997, and will be followed by a second. The freezing or reductions of emissions agreed under the first protocol are not yet fulfilled for all signatory States. The next protocol will require more extensive reductions based mainly on the needs for a decrease of the nitrogen load on ecosystems. NO_x reductions must be achieved in urban as well as in rural areas in order to reduce total emissions. These reductions will also be beneficial for reduction of the NO₂ concentrations in urban air.

In most urban areas traffic is a dominant contributor to ambient air concentrations of NO₂. Control measures on traffic are therefore of great importance for reduction of exceedances of health related limit values. For roadside concentrations, traffic planning may be sufficient to reduce exceedances of short-term limit values. To reduce long-term average concentrations in urban background and rural areas, it is necessary to reduce the specific emissions and/or the traffic. However, there are areas, where the contribution from point sources may be as important. Control measures for point source emissions also need to be considered, especially for protection of the ecosystems.

To achieve efficient reduction strategies for ambient air, the atmospheric chemical processes must be considered. O₃ and other oxidants may lead to relatively ineffective reductions of NO₂ concentration, even if the emissions of nitrogen oxides are substantially reduced. Model calculations used for studying different emission scenarios, must involve sufficiently detailed atmospheric chemistry.

Reduction of NO_x emissions will be achieved by measures already planned or decided. One recent study - Energy 2020 suggests that as a result of current EU and national legislation NO_x emissions in the EU-15 will drop by 20% in the year 2000 and by 33% in the year 2010 (Capros et al., 1995) (compared to 1990). This is based on economic growth and traffic volume assumptions in the conventional wisdom scenario. A second study indicates that with current legislation, including the proposed EU Directive on off-road vehicles as well as current national reduction plans (the mild scenario), NO_x emissions might fall by around 30% in the year 2010 (compared to 1990). NO_x emissions from non EU-countries in Europe are expected to drop by roughly 10% over the 1990 levels in the year 2010 (Commissions Services, 1995). Further planned EU legislation might reduce NO_x emissions by 53% in the year 2010. According to a third study current legislation might be sufficient to reduce NO_x emissions by 47% in the year 2010 (Cofala et al., 1995). The different results of these studies arise from differences in energy demand, transport forecast and pollution control implementation assumptions.

The emission reductions expected vary from country to country. Table 4.1 summarises the emission reductions of the conventional scenario and the acidification study.

Table 4.1 Expected reductions with current legislation (% over 1990). Required reductions are the estimates given in annex IV.

	Conventional Wisdom	Acidification (mild scenario)	Required reductions (Annex IV)
Austria	25	35	50
Belgium	34	30	5-45
Germany	40	31	10-30
Denmark	29	35	
Finland	31	32	
France	25	34	
Greece	15	18	
Ireland	17	29	
Italy	32	32	
Luxembourg	32	26	
Netherlands	40	78	43
Portugal	-6 (increase)	46	5-40
Sweden	31	39	10-20
Spain	19	29	
United Kingdom	42	40	

The little evidence available suggests that the reductions in NO_x emissions expected by the year 2010 might, apart from specific local problems, meet limit values in Belgium, Germany, Sweden, Denmark and perhaps the Netherlands. Problems are likely to persist in Portugal. Further analysis is required, however, since many countries the required additional reductions compared to current legislation, are not

known. In addition, the impact of current legislation needs further examination to resolve results obtained by different studies.

In addition to EU vehicle and stationary source regulations many national initiatives to reduce NO_x emissions exist, or are planned. The impact of these should be considered.

4.2 Techniques to reduce ambient air concentrations of NO_2

The NO_2 concentrations in different scales

The concentration of NO_2 in all temporal and spatial scales is determined by the total emission in the region, dispersion of pollutants, deposition of NO_2 , many complicated chemical transformations and by the long range transport of nitrogen compounds.

In rural areas, far from air pollution sources the temporal and spatial variations of the NO_2 concentrations are relatively small. In these areas the limit value for protection of vegetation is important, i.e. the annual average concentration. However, for protection of vegetation, deposition in relation to critical load will often be even more critical. There is no simple linear relationship between the emission of nitrogen compounds between regions and the concentration of NO_2 in the air. However, advanced large scale models such as the EMEP model, based on data from European emission inventories, meteorological data and the atmospheric chemical and physical processes, have been established and can be used to estimate the concentrations of NO_2 (and O_3) in rural areas (Barrett et al. 1995), (Simpson 1992), (Simpson 1993).

In urban background areas where local sources have a more direct impact on the NO_2 concentration, no linear relationship between the emission of nitrogen oxides and the NO_2 concentration here either. Urban background is often representative for residential areas and thus for human exposure; the long term as well as the short term limit values may be critical.

At local sites, e.g. in streets, the NO_2 pollution is governed partly by the local emission of NO and NO_2 . It is important again to emphasis the non-linearity of the governing chemistry for NO_2 . During most periods NO_2 levels near urban streets are limited by the ozone levels and not by the NO_x releases in the street. This means that the very local scale phenomenon of high NO_2 concentrations in a street is coupled to large scale phenomena, since high ozone concentrations are caused by long range transport. However, in streets, short term peak concentrations may occur and here the short term limit value for protection of health is the most critical.

Assessment of present status and need for abatement - necessary tools

In order to construct a model tool for determining the effects of different control strategies, local scale dispersion and chemistry of nitrogen compounds and ozone, as well as long range transport of ozone (and to some extend NO_x) must be taken into account. The only practical way to construct such a tool is to combine a transport-chemistry model for long range transport to a local scale model for a single street. However, the long range transport model also has to be able to account for the dispersion and chemistry on an urban scale of a few kilometres.

Such a model has been developed at the National Environmental Research Institute, Denmark. In this case the variable scale Atmospheric Chemistry and Deposition (ACDEP) model is used for the long range transport and dispersion inside the urban area. The results from this model are used as input to the Operational Street Pollution Model (OSPM) for dispersion in a single street. OSPM accounts for the chemical formation of NO₂ in the street (*Berkowicz, 1997*).

A modelling tool of this kind is necessary to study the result of different control strategies on both pollution levels and, when coupled to an exposure model, on population exposure to traffic air pollution.

However, a total assessment of the cost and benefits of the measures to be taken must be based on integrated social-economic models, which in principles should include all relevant aspects for the society.

Different approaches to reduction of pollution levels for NO₂

There are a number of possibilities to achieved reduced NO₂ concentrations; many different actions such as legislation on emission reductions of NO_x, initiatives for a complete change of infrastructure and behaviour of the society. These actions can be taken on different scales from the EU level over national decisions to local initiatives. Table 4.2 contains actions suggested by the Working Group. The table is not complete, other actions may also be relevant .

Table 4.2 Actions for reduction of the NO₂ concentrations in air suggested by the Working Group. Other actions may also be considered.

EU level	National level	Local level
Vehicle emission reduction	Emission standards	Local vehicle restrictions
Vehicle inspection	Environmental permit	Alternative fuels
Energy conservation	Energy conservation	Energy conservation
Point source emission reductions		
Fuel standards		
Air quality standards	Transport infra structure (social, land-use etc.)	Alternative public and private transports
Fiscal incentives	Fiscal incentives	Road-, parking-pricing
Sustainable city planning	Informatic highways	Traffic planning, management
Behaviour, education, awareness	Behaviour, education, awareness	Behaviour, education, awareness
Car fleet renewal (scrapping subsidies)	Traffic planning	Transport system planning
Traffic planning		Traffic information
		Environmental zoning
Research	Research	

Emission reduction possibilities

The emission of NO_x and other pollutants important for the formation of NO₂ can be reduced by energy saving, reduction of activities, alternative fuels, improved combustion technology, scrubbers, catalysts etc. They include technical as well as non-technical measures. The most cost-effective reductions are often obtained by a combination of the different methods. The abatement strategy should be decided on an overall evaluation of the cost for a certain reduction leading to decreased NO₂ concentration. In addition, these considerations should be co-ordinated with other measures taken for the reduction of other pollutants. The following are examples on relevant considerations in connection with emission reductions.

Road traffic the main contributor to the NO_x-emission in Europe. For large Auto-Oil Programme, has been carried out as a co-operation between the European, and the Commission European Motor and Oil Industry.

The aims of the study were:

- To quantify further reductions in road traffic emissions which may be needed to achieve agreed air quality objectives.
- To identify sets of measures - including improvements in fuel/vehicle technologies as well as other measures such as inspection and maintenance programmes, traffic control - that can achieve such reductions.
- To evaluate them from a cost/effectiveness point of view and to search for the most cost-effective proposals.

The results of the programme confirm that both fuels and engine technologies are important determinants of motor vehicle emissions. Relationships between fuel properties, engine technology and exhaust emissions exist. Based on fuel parameters and vehicle technology studies the programme was able to quantify these relationships in tables and equations to be used as input for air quality and cost optimisation models.

Changes in a given fuel property may lower the emission of one pollutant but may increase those of others. In some cases, engines in different categories, such as heavy duty and light duty vehicles, have disparate responses to changes in fuel properties. Thus, no simple answers exist but the basis for the most cost-effective combination in terms of improved vehicle technology and improved fuel properties may be identified.

The cost effectiveness methodology and study was developed by consultants to the commission, using cost data provided by the Oil and Motor industries. It is intended to provide the basis for the Commission's proposal for post 2000 vehicle exhaust emission requirements, and should also be used in this exercise.

Based on the NO_x-protocol, decisions on reduction of the NO_x-emission in the European countries have been taken. Cost estimates for technical measures to control NO_x emissions from stationary and mobile sources are currently available (Amman and Klaasen, 1995). Problems in estimating cost might occur for non-technical

measures such as road pricing, since data are scarce. More cost estimates will be made for different source categories in the different countries. This will also include non-traffic sources, e.g. power plants, space heating systems, industrial plants etc. These estimates can be used as an input for evaluation of the cost implications of the new NO₂ -limit values.

4.3 Benefits by the reduction of NO₂

The limit values recommended in section 2 are proposed with the primary aim of protecting human health. However, by reducing ambient air concentration of NO₂ other benefits will be achieved such as reductions in material damage and improvements in ecosystems. In urban areas the number of people exposed to high peak concentrations and to high average exposures will decrease. Information on health benefits are to expected to be estimated by WHO.

As seen from the risk assessment, there is no exposure - effect relationship for NO₂ and materials available today. Consequently, there is insufficient evidence to quantify the magnitude of damage (e.g. of cultural heritage) and thus the benefit of reductions (as discussed by Mohr et al., 1997) .

Despite our inability to evaluate the benefits in a quantitative way, it is possible to list the benefits qualitatively. Under conditions of reduced NO₂ levels in urban atmosphere, a lower rate of accumulation of nitrate by building surfaces would be expected, which in turn, should lead to less corrosion by HNO₃ and limit any role that nitrogen oxides might play in the oxidation of SO₂ to H₂SO₄ on the urban fabric. Secondly a lower rate of direct damage would be expected to sensitive materials such as paper and cloth from nitrogen oxides indoors. Finally, less nitrogen would be available as a nutrient to micro-flora and thus lower rates of biological growth and disfigurement of buildings could be expected. Reduction of emissions of NO_x from traffic and point sources will also result in lower NO₂ concentrations in rural areas. Even in areas, where no exceedances are seen of the limit value to protect vegetation, there is a beneficial effect due to decreased deposition. Quantification of effects of NO₂ and nitrogen deposition, acidification as well as eutrophication of lakes, the sea, crops and ecosystems are dealt with within the mapping of critical loads and levels. This was carried out to be a basis for the negotiations on NO_x emissions reductions within the new NO_x protocol. In the Integrated Assessment Modelling however, only abatement costs are included and not the benefits of reduction. Results from Integrated Assessment Modelling within UNECE in connection with the second NO_x protocol is expected to be available this summer.

Further benefits will be achieved if the emission reductions are large enough to result in decreased ozone formation. Since the effect of reductions may vary from one part of Europe to another, due to chemical characteristics of the local atmosphere, it is difficult to find optimal abatement strategies. Quantification of the effect on ozone formation in different areas as a consequence of various reduction scenarios is studied within the EMEP modelling activities as a basis for the new NO_x protocol.

Finally, abatement strategies, chosen with the aim of reducing NO₂ concentrations in ambient air, may be performed with techniques that lead to reduction of other air pollutants and, in this way, also to reduction of other environmental effects. Such additional effects may, however, be difficult to quantify, unless the techniques for reduction are known in detail.

4.4 Costs of lack of action

The cost for lack of action includes the cost for existing damages. The cost for lack of action are, in principles the benefits foregone or the benefits that could have been obtained, if action was undertaken. These benefits could consist of decreased morbidity, i.e. respiratory illness, and reductions in the damage to buildings and ecosystems. These also include an estimate of the situation if no actions are taken, i.e. business as usual. The development of the economy necessitates increases in traffic, transport, energy use, industrial production etc..

The costs of NO₂ damage to health and the environment are more difficult to estimate than e.g. the costs associated with SO₂ effects on materials and crop loss due to O₃ exposure. This is due to the fact that NO₂ effects have been difficult to separate from effects due to SO₂ and O₃.

One Europe wide study assessed the costs of health damage due to NO₂ concentrations on human health (Ozdemiroglu and Pearce, 1995). This study took the work of the WHO, cited in chapter 2, as the basis. The WHO study suggested that at least 21 million people, living in urban areas, were, at the end of the eighties, exposed to NO₂ concentrations exceeding the daily WHO AQG (of 1987) (UNECE, 1995). The most likely effect on health associated with these elevated concentrations in an increase in respiratory symptom requiring medical visits. the WHO study indicated that for the cities where air quality data were available, 17000 to 19000 incidences per year of LRI (Lower Respiratory Illness) would occur. Assuming a similar exposure pattern across Europe (East and West) as in the cities with data, 58000 to 99000 incidences would occur. Ozdemiroglu and Pearce (1995) translated these physical impacts in monetary terms on the basis of a review of studies that analysed the willingness-to-pay of people to avoid an emergency room visit or a symptom day. A mild symptom day was valued a £ 5 and a severe one, requiring a medical visit, at £ 155. The results are summarised in Table 4.3.

The long term effects of increased NO₂ concentrations in the form of decreased lung functions are not taken into account in the above study. The WHO estimated that 17 to 60 million people might suffer from a decrease in lung function of 2 to 5 percent. No estimates are available of the associated costs.

Furthermore, NO_x emissions indirectly contribute to ground level ozone. Any reductions in NO_x emissions to meet NO₂ limit values might then indirectly lead to additional benefits; or reduced damage costs, due to reductions in ozone concentrations. Ozdemiroglu and Pearce (1995) state that the short term health effects due to elevated ozone levels lead to 0,5 to 4 million cases of throat and eye irritation

in children per year. Using a £5 estimate for a mild symptom day they estimate the damage costs at £2.3 to £20 million. The willingness-to-pay for avoiding this damage might be higher in reality because of altruistic motives.

Table 4.3 Annual Costs of lower respiratory illness due to NO₂

Area	Number of incidences	Total costs (£ Sterling)
Selected cities	17,000 - 29,000	mild: 85.000 - 145.000 severe: 2.6 - 4.5 million
The whole of Europe	58,000 - 99,000	mild: 290.000 - 495.000 severe: 9 - 15 million

Costs for lack of action can be calculated from the effect quantification in 4.3. At present we have no data at all on cost for damage due to NO₂. We believe it is possible to estimate the costs of health effects; hospital care for certain diseases, costs for medicine, cost for absence due to sickness. In a Swedish study (Leksell & Löfgren 1995) using the Contingent Valuation Method, CVM, for estimating costs concluded that the damage from inhaling various pollutants could be estimated at \$0.50 per milligram inhaled. It was seen in the interview study, included in the Swedish cost study, that people put a higher price on pollution in city centres than in the outskirts or the countryside. The higher the concentrations, the more value they were inclined to put on a reduction. The price put on emissions of NO_x and VOCs in Gothenburg was \$6.00 per kg. In the city centres, where the exposure was highest, a ten times higher price was set: \$60 per kg.

Cost for damages caused on cultural heritage and materials. Some data may be published in the report from the UN ECE workshop on Economic Evaluation of Air Pollution Abatement and Damage to Buildings including cultural heritage, 23 - 25 January 1996. However, the report is not yet published. Estimates of total corrosion costs have been estimated to between \$ 3.40 and \$26 per inhabitant per year, equal to between \$2 and \$18 billion for Europe as a whole. The largest part of this is however due to SO₂.

Over Europe there are national studies made in many countries, Norway (Central Bureau of Statistics) and Sweden (Central Bureau of Statistics and National Institute of Economic Research). So far sulphur is mainly studied, but work is proceeding for NO_x.

It may be possible to estimate NO₂ effects on Vegetation from the mapping of exceedances of critical levels. The estimation of ecosystems damage due to oxidised nitrogen deposition may be possible from the exceedance of critical loads. However, the effects of NO_x deposition must be dealt with in relation to the deposition of NO_x and sulphur, and the relevant costs may be difficult to estimate. The value of cultural heritage and ecosystems is not easily defined with traditional economic methods.

4.5 Discussions and conclusions

The present ambient air concentrations exceed the recommended limit values in many parts of the EU and large reductions may be necessary in some areas, if every location is to comply with these values. Other areas within the EU either comply with the limit values, or the exceedances are small.

A preliminary list of effects, suggested by the Working Group, for which cost-benefits estimates should be estimated is shown in table 4.4.

Table 4.4 Effects to be considered in the cost benefit analysis. The list should be supplemented and prioritised.

Health	Ecosystems	Material	Amenity
Acute health effects	Acidification	Corrosion	Visibility
Chronic health effects	Crops loss/gain	Coating	Tourism
Hospital admissions	Forest damage/gain	Soiling	Well being
Mortality	Eutrophication	Changes of colour and structure of surfaces	
Morbidity	Diversity	Chemical weathering	
Medication		Cultural heritage	
Work loss			
School loss			
Reduced-Activity-Days; RAD			
Clean air resort treatment			

The present situation varies significantly between the different Member States, which means that the starting point differs. In addition the priority of effects are different within different EU regions. For example, the effects on ecosystems may be most important in the north and the health effects more important in the southern part of Europe. It has to be emphasised that the adverse effects, costs and benefits and the possible actions against pollution for the different pollutants are closely linked and therefore multi-pollutant, multi-effect perspectives are necessary.

Because the limit values based on effects will be exceeded in many areas of the EU it should be decided, which levels of effects are acceptable and for how long. However, it is recommended that the time of compliance should be based on the least cost time schedule. An estimate of the sensitivity of the time schedule should be made, e.g. by cost estimates on different scenarios.

The cost study should comprise all costs, including also all external costs. As far as possible, a uniform basis for the cost estimates is wanted but, when significant, differences between the Member States must be taken into account.

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Annex I

Guidelines given for NO₂ in order to protect health and the environment in European countries.

(Concentration is given in Tg/m³ (ppb in brackets))

Country	Mean value for time period	Conc.	Comments
Austria	30 min.	200 (110)	Guide line
	Daily	100 (50)	Guide line
Austria (lower Austria and Styria)	30 min.	200 (110)	Limit value
Austria (upper Austria)	Daily	100 (50)	Limit value
	30 min.	300 (160)	Limit value
	Daily	100 (50)	Limit value
	97.5 percentile of 30 min means	150 (80)	Limit value
	Monthly	50 (25)	Limit value
Belgium	98%-percentile 1-hour mean	200	Calculated over one year
Bulgaria	30-min. mean	85 (45)	Protected areas
Czechoslovakia	Daily mean	100 (NO _x)	
	½-h mean	100 (NO _x)	
EU	98%-percentile	200 (110)	1-h means over a year or shorter time periods.
			Limit value to protect human health and environment.
	50% (Median)	50 (25)	Guideline to improve the health protection and term for long protection of environment.
	98%-percentile	135 (75)	Same as above. 1-h means over a year or shorter time periods.
Finland	Daily mean	70 (37) 1996	
	99-percentile (1 h) per month	150 (..80)	Guideline, 99-percentile
France	98-percentile (1 h)	200 (105)	Limit value
	95-percentile of 24-h means	200 (110)	Limit values for estimating the stack heights.

Germany	Yearly mean	80	To protect health
	98-percentile of 30-min. means	200	To protect health
Hungary	Yearly mean	50 (25)	Stated 1.1. 1987.
	24-h mean	70 (35)	Especially protected areas.
	30-min. mean	85(45)	
	Yearly mean	70(35)	Protected areas.
	24-h mean	85(45)	
	30-min. mean	100 (50)	
	Yearly mean	120 (65)	
	24-h mean	150 (80)	
	30-min. mean	200 (110)	
Italy	1-hour mean	200 (110)	Stated of the National Health Board (Jan. 1982) to protect health. To be exceeded not more than once a day.
Liechtenstein			Limit values equal to Switzerland of 1986.
Luxembourg	98-percentile	200 (NO _x)	
	of daily means		
The Netherlands	98-percentile of 1-h means.	135 (66)	Limit value
	99.5-percentile, 1-h means.	175 (87)	Limit value
	50-percentile, 1-h means.	25 (12)	Guide value
	98-percentile, 1-h means	80 (40)	Guide value
Poland	30-min. mean	150 (NO _x)	Especially protected areas.
	Daily mean	50 (NO _x)	
	Yearly mean	8 (NO _x)	
	30-min. mean	500 (NO _x)	Protected areas
	Daily mean	150 (NO _x)	
	Yearly mean	22 (NO _x)	
Portugal	98-percentile	200 (NO _x)	short-time values are 1-h means over a year or a shorter period.
Romania	30-min. means	300 (160)	
	Daily means	100 (50)	
Sweden	98-percentile of 1-h means.	110	Limit value.

	98-percentile of 24-h means.	75	To protect human health.
	Means from Oct. to March.	50 (25)	Same as above.
Switzerland	95% of ½-h means of one year soil from pollution; used both at stationary plants and in urban environment; measured at separate sampling stations.	100 (50)	To protect people, animals, and vegetation as well as
	24-h means	80 (40)	To be exceeded not more than once a year.
	Yearly mean	30 (15)	
Spain	98-percentile	200 (NO _x)	Limit value
	98-percentile	135 (NO _x)	Guideline
USSR (former Soviet Union)	Daily mean	40 (NO _x)	
	20-min. mean	85 (NO _x)	

Annex II

Guidelines given for NO₂ and NO in order to protect vegetation in European countries.

Guidelines or limit value defined by:	Level	Tg/m;	Mean value for time period	Comments
Bad Harzburg (1988)	30		Annual mean	Over all vegetation type
Guiderian (1988)	20		Annual mean	
	40		Winter mean	
	60		Growth season mean	
	10			In order to protect eutrophication poor vegetation
	800		1 hour mean	
The Netherlands (1982)	95		4 hour mean	Recommended by the Dutch Health Council in order to protect vegetation
WHO (1994)	30		Annual average	Sum of NO and NO ₂ , added in ppb and expressed as NO ₂
EU (1986)	30		Annual mean	
	10		Annual mean	In order to protect eutrophication poor vegetation
VDI (1978)	600		30-min. mean	In order to protect sensitive vegetation
	350		Growth season mean	
Austria (1988)	200		30-min. mean	In order to protect vegetation
	80		Daily mean	
	30		Annual mean	
	80		30-min. mean	In order to protect highly sensitive plants
	40		Daily mean	
	10		Annual mean	
Finland (1996)	30		Annual mean	Guideline, sum of NO and NO ₂ (as NO ₂)

Annex III

NO₂ measurement, sampling and calibration methods

NO₂ MEASUREMENT METHODS

Methods	Description	Referen	Advantages/disadvantages
1. Manual methods			+ cost effective - discontinuous and time consuming measurements
1.1. Modified Griess-Saltzman	Bubbling in absorbing solution (azo-dye forming reagent) + colorimetry	ISO 6768	- possible interferences - uncertainty of "Saltzman factor"
1.2. Saltzman	Bubbling in absorbing solution (azo-dye forming reagent) + colorimetry	VDI 2453 Bl.1	- possible interferences
1.3. Arsenite	Bubbling in absorbing solution (NaOH/NaAsO ₂) + colorimetry	-	- possible interferences
1.4 Diffusive sampling	Diffusive sampling onto absorbent (TEA) + colorimetry	-	+ cost effective, ideal for large scale monitoring + continuous, real time measurements - requires regular calibration and maintenance
2. Automated methods			
2.1. Coulometric	Redox reactions in electrolytical cell + electric current measurements	-	- possible interferences
2.2. Chemiluminescence	Conversion of NO ₂ to NO + measurements of chemiluminescence reaction between NO and O ₃	ISO 7996	+ high linearity, currently most commonly used method in networks
3. Optical methods			+ simultaneous multi-component analysis - integrated concentration over path length
3.1. DOAS	Differential optical absorption along path length	-	+ easy, maintenance-free operation - expensive analyser
3.2. DIAL	Differential optical absorption of backscattered laser beam		+ 2D-mapping possible - expensive analyser

NO₂ CALIBRATION METHODS

Methods	Description	Referen	Advantages/disadvantages
1. Permeation method	NO ₂ permeation through a membrane into a flow of carrier gas, at constant temperature + periodic or continuous determination of NO ₂ losses by weighing.	ISO 6349	<ul style="list-style-type: none"> + primary calibration method + diffusion rate determined by weighing + continuous production of calibration gas mixture + cost effective method + precise and accurate (uncertainty $\pm 5\%$) - control of NO₂ purity required
2. Static volumetric method	A known volume of NO is added to a known volume of complementary gas, under controlled pressure and temperature conditions. Combined with gas phase titration for the determination of the analyser converter efficiency.	ISO 6144	<ul style="list-style-type: none"> + primary calibration method + cost effective method + also suited for other pollutants + precise and accurate (uncertainty $\pm 3\%$) - difficult handling - control of NO purity required
3. Gravimetric method (high or low concentration mixtures)	A chamber is weighed before and after introduction of a certain quantity of NO and NO ₂ , then filled up with air and pressurised	ISO 6142	<ul style="list-style-type: none"> + gas cylinders commercially available + easy handling - poor stability of concentration mixtures with time - unknown accuracy - no primary calibration method
4. Dynamic volumetric method	Introduction of a given rate of a gas into a constant flow rate of a complementary gas. The gas is usually a high concentration gas mixture obtained by the gravimetric method.	ISO 6145	<ul style="list-style-type: none"> + gas cylinders commercially available + easy handling - unknown accuracy - no primary calibration method
5. Gas phase titration	Dynamic diluted NO reacts with a known concentration of O ₃ to NO ₂	EPA	<ul style="list-style-type: none"> + easy handling - no primary calibration method

NO₂ SAMPLING METHODS

Methods	Description	Referen	Advantages/disadvantages
1. Laminar flow manifold	Flow 150 l/min, tube diameter 15 cm. Inlet material: glass, stainless steel, Teflon	EPA	<ul style="list-style-type: none"> + isokinetic sampling, sample unaffected
2. Turbulent flow manifold	Modular sugar cane design. Inlet material: glass, stainless steel, Teflon	-	<ul style="list-style-type: none"> + low cost, modular construction
3. Sampling without manifold	Direct connection of analyser inlet to station sampling head	-	<ul style="list-style-type: none"> + low cost, efficient sampling without losses

Annex IV

Rough estimate of the need for reduction of NO₂ concentrations in urban areas in different Member States

(Answers to questions sent out to Steering Committee representatives)

	Present situation; exceedances of limit values.	Estimated need for reduction of NO₂ concentration to comply with the recommendations (%)	Estimated need for NO_x emission reduction (%)
Austria	STLE and LTLE in major cities near heavy traffic streets. VLE not in forest and agriculture areas.	About 50% is estimated at some "hot spots".	traffic and dom. heat. ?
Belgium 1995	STLE 1 area LTLE 3 areas	4% 30 - 45% roadside 5 - 18% other stations	? Red. of traffic NO _x necessary.
Finland	STLE normally not exceeded, but in episodes in 1995 in some major cities. LTLE exceeded before 1995 (but decreasing) VLE not exceeded		
Luxembourg 1992 - 95	STLE No exc. LTLE 2 stations VLE 3 urban station. No at rural stations.	Not possible to estimate, no evaluation made.	
Portugal	STLE 5 urban and 2 industry areas. LTLE 4 urban areas. VLE in 4 urban areas	20 - 40% 5 - 25% 25 - 50%	Car emissions Red. of car em in cities.
Germany	STLE and LTLE nearly all stations with intense traffic and most traffic related urban stations.	In general 10 - 30% At some hot spots >30%	Roughly at least 25-30 %. Add need at hot spots
Netherlands	STLE frequent LTLE many cities	43% is needed, in add. to pres. policy 30% is needed.	corresponding to 47% NO _x , more in busy streets.
Norway, measurements during winter	LTLE in Oslo, 8 cities with 50 - 60 as winter av.	No evaluation	? No evaluation.
Sweden, measurements during winter. 94/95	STLE 1 station LTLE in large cities near traffic.	approx. 10% approx. 20%	? No evaluation. Mild winter may influence.

STLE is Short Term Limit Value Exceedance. LTLE is Long Term Limit Value Exceedance. VLE is Vegetation Limit Value Exceedance.

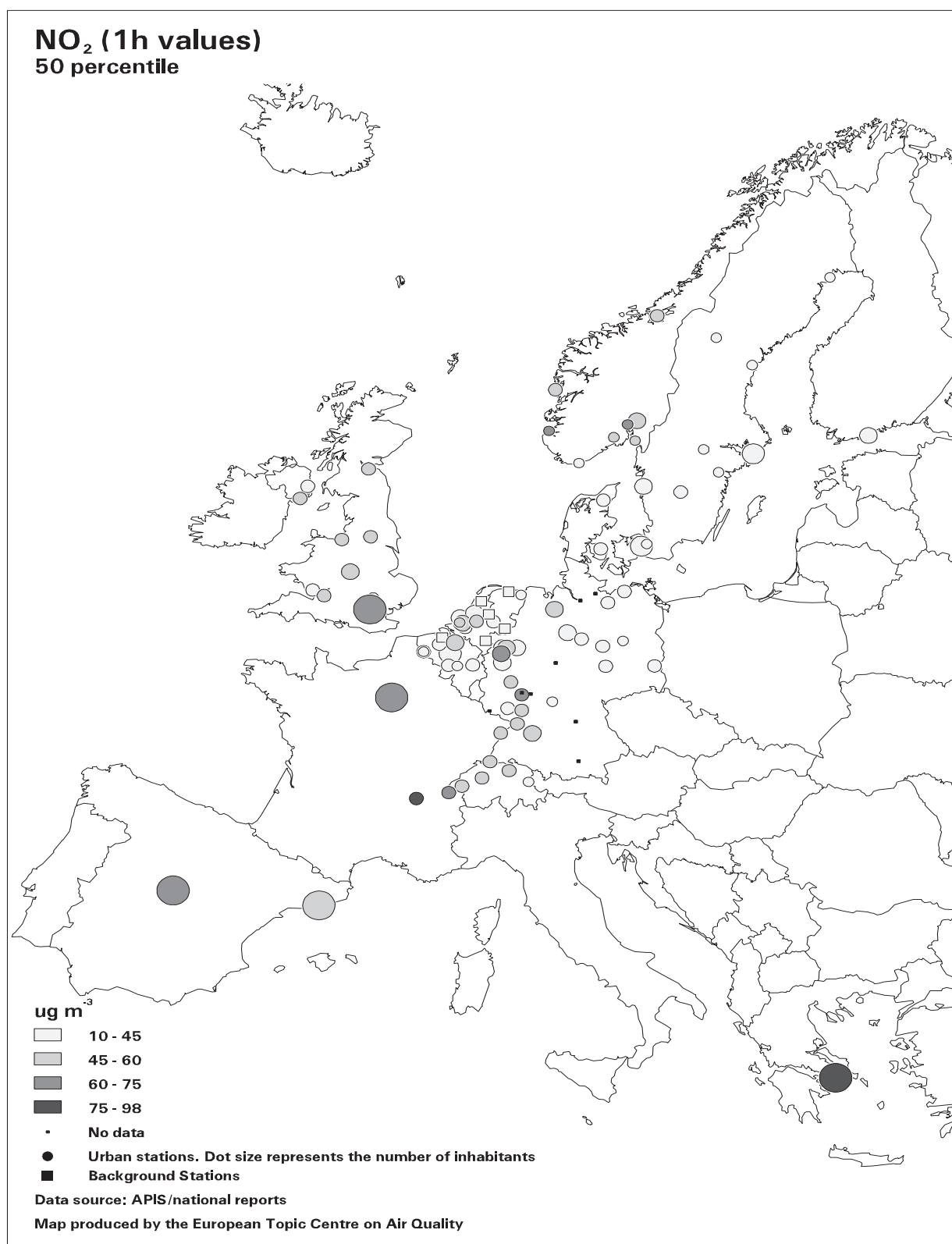
Annex V

Concentration levels at sites exceeding guidelines for NO₂ in Europe, 1993. Based upon all data available to the topic centre.op

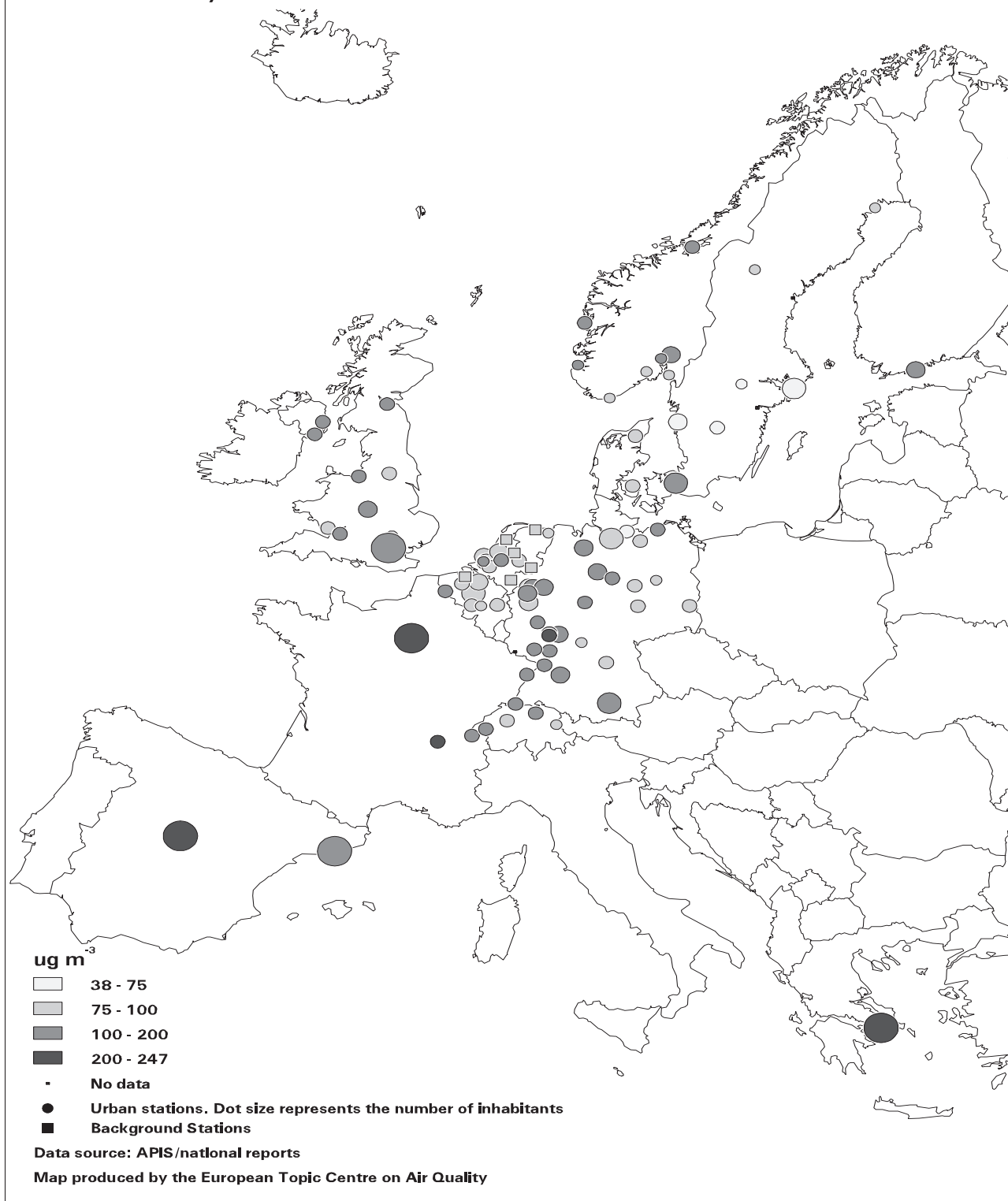
		EU Guide Values		WHO Guideline Values	
		Median	P 98	Max. 1h	Max. 24h
		>50	>135	>400	>150
Germany					
Ludwigshafen-Frankental	Mundenheim	51			
	Frankental	57			
Mainz-Mombach	Parcusstrasse	67			
Koblenz	Friedrich-Ebert-Ring	57			
Karlsruhe	Karlsruhe-Mitte	51			
Wiesbaden	W-Ringkirche		136		
Essen	E-Ost	53			
Hannover	Göttinger Strasse		136		
Greece					
Athens	Patission 147	98	247	554	253
	Pireas Platis Dimotikou	67	136		
Norway (based upon 24h values)					
Oslo	St. Olav Square	50			
Bergen	CMI	59	135		
Trondheim	Torget	57			
Stavanger	Handelens hus	63			
Drammen	Engene	74			
Frederiksstad	Broch street	50			
Skien	King street	57			
Spain					
Madrid	Cuatro Caminos	72	187	448	184
	Carlos V	57	151		154
	Plaza Espana	57	187	470	189
	Plaza Castilla	58	141		
	Villaverde	72	151		
	Arturo Soria	53	159		
Switzerland (mean values)					
Basel	Feldbergstrasse	57			
Bern	Bern	57			
Genève	Ile	60			
Lausanne	Lausanne	50			
	Rue Cesar Roux 2	56			
Zürich	Schimmelstrasse	57			
The Netherlands					
Utrecht	Wittevrouwenstraat	60			
United Kingdom					
London	Bridge Place	63	136		192
	London Bloombury	65			
Edinburg	Edinburg centre	50			
Newcastle	Newcastle centre	54			
Leeds	Leeds centre	50			
Liverpool	Liverpool centre	50			

Annex VI

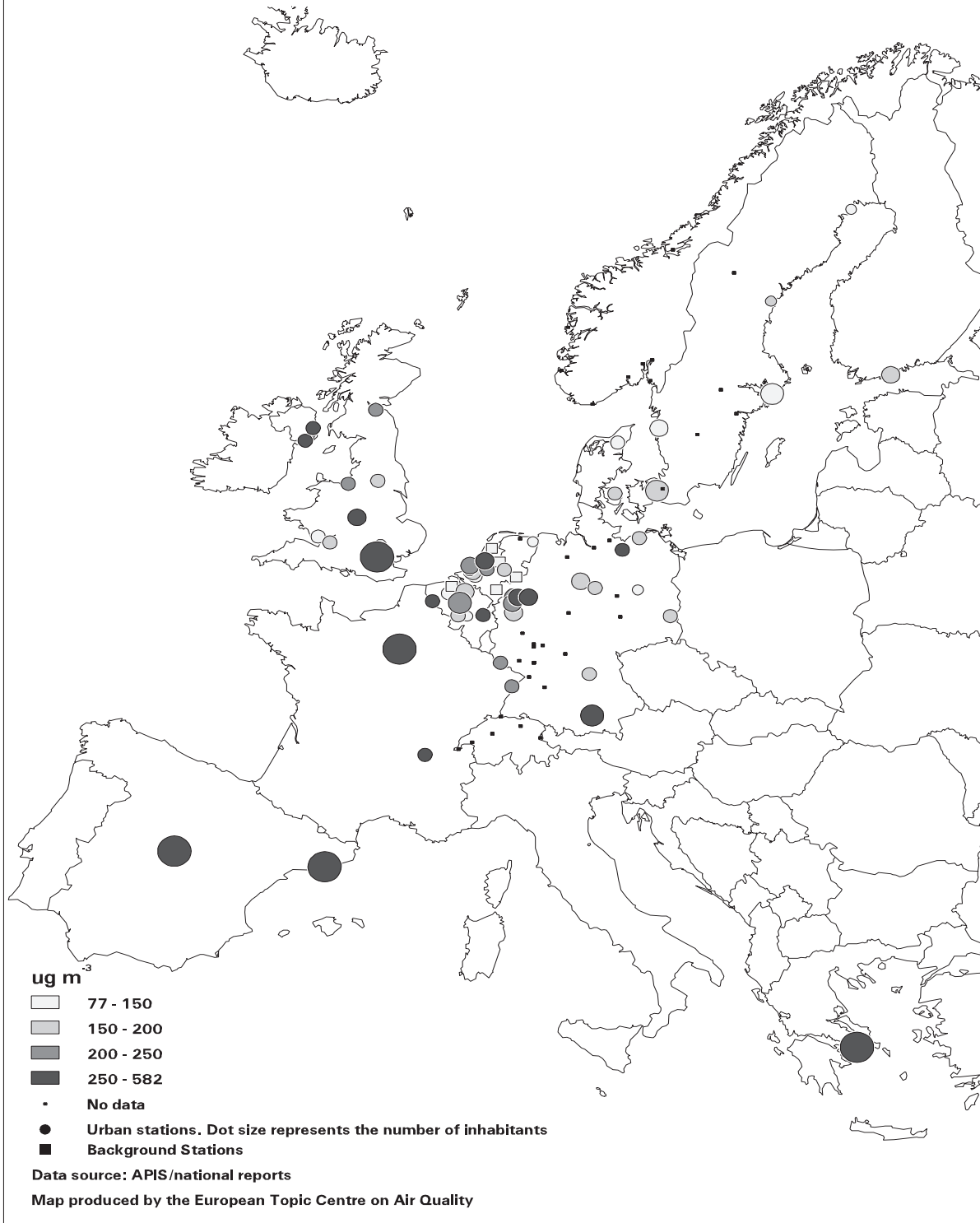
Measured NO₂ concentrations in Europe.



NO₂ 98 Percentile Based on hourly values



NO₂ (1h values) **Maximum hourly concentration**



Annex VII

Harmonisation and normalisation of models

There are various harmonisation and normalisation efforts are in progress. A description of activities is made by (Olesen,1995).

Activities within EC DG XI and DG XII

In the work of DGXI in preparation of a framework directive on air quality and in some fuel specification studies, the modelling aspect is considered. Two of the research programmes of the DGXII Framework Programme are related to short-range modelling. DGXII supported the Model Evaluation Group (MEG) in the evaluation of technical models used within the major industrial hazard zone. Guidelines for model developers have been issued (MEG 1994).

EEA ETC-AQ

Project MA3 "Harmonisation of the use of models" includes co-operation with the European ad-hoc initiative for harmonisation within atmospheric dispersion modelling for regulatory purposes. Three sub-projects deal with the use of models:

Project MA3-1 Review requirements on models and model application. The review is based on Europe's environment - the Dobbris assessment, a questionnaire sent to 38 European countries, and requirements and needs from international organisations (de Leeuw et al., 1995).

Project MA3-2 Report on state of the art: needs and trends. The report by Moussiopoulos et al. (1995) contains state of the art for models on various scales: global, regional-to-continental, local-to-regional and local. Quality assurance of models and trends in air pollution modelling.

Project MA3-3 Establish documentation centre and tool-kits for testing relevant models. The first reports from the sub-project are scheduled to 1996.

Project MA3-4 Report on guidance in selected zones and in application of models. The first reports are scheduled to 1996.

COST

Three COST actions are directed towards modelling aspect. CITAIR contains a database, monitoring networks and modelling of urban air pollution. The COST technical Committee on Meteorology is starting action on harmonisation of meteorological pre-processors. The COST action F1 is aiming at prediction, modelling, manipulation and control of complex three-dimensional viscous flows.

Ad-hoc initiative for harmonisation in atmospheric dispersion modelling for regulatory purposes

A series of workshops have become a meeting place for increased co-operation and harmonisation of atmospheric dispersion models for regulatory purposes (Cosemans,

1995). A fourth workshop will be held during spring 1996 in Oostende to deal with: Further experiences with and extension of model validation kits (such as data sets of SF₆ dispersions experiments), model output inter-comparison exercises, harmonisation in the pre-processing of meteorological data for dispersion models, regulatory models in specific countries and practical application of atmospheric transport and dispersion models.

Nordic countries

A group of Nordic modellers has evaluated the utilisation of models to assess air quality in cities from a Nordic perspective. The evaluation includes an overview of models for different scales used in the Nordic countries, with the emphasis on street/road and urban scale models (Larssen et al., 1995).

National work

Some work is being made nationally. Most of it is done in the US. The Royal Meteorological Society in UK has issued a set of guidelines on the choice and use of models within atmospheric dispersion (Royal Meteorological Society, 1995). The Dutch Ministry on Housing, Physical planning and Environment has issued a Dutch standard on the description of air pollution models (Noordijk, 1992; Dekker and Sliggers 1992; Dutch standard on Description of air pollution models, 1992).

Standardisation

The International Standardisation Organisation (ISO) has established a subcommittee on Meteorology to the Technical Committee on Air quality. The European Committee for Standardisation (CEN) is not presently dealing with modelling.