

AMBIENT AIR POLLUTION BY PARTICULATE MATTER

POSITION PAPER

EXECUTIVE SUMMARY

Descargado desde:



This paper was prepared by the Technical Working Group on Particles. This Group was set up by the European Commission to help it prepare a daughter directive on ambient air particle pollution in the context of the Ambient Air Quality Assessment and Management Directive (96/62/EC). It consisted of experts from Denmark, France, Germany (co-chair), the Netherlands, Spain, the United Kingdom (co-chair), the Commission (DGXI Environment and DGXII Research), UNICE (Union of Industrial and Employers Confederations of Europe), the European Environment Bureau, the European Environment Agency, the Joint Research Centre (Ispra) and the World Health Organization.

1. POLLUTANT DESCRIPTION

Airborne suspended particulate matter (PM) can be either primary or secondary in nature. Primary particles are emitted directly into the atmosphere either by natural or anthropogenic processes, whereas secondary particles have a predominantly man made origin and are formed in the atmosphere from the oxidation and subsequent reactions of sulphur dioxide, nitrogen oxides and VOCs. In most European countries, industrialisation and high volumes of traffic mean that anthropogenic sources predominate, especially in urban areas, and sources of anthropogenic particles are similar throughout Europe. The most significant of these are traffic, power plants, combustion sources (industrial and residential), industrial fugitive dust, loading/unloading of bulk goods, mining activities, human-started forest fires and, in some local cases, non-combustion sources such as building construction and quarrying. The main natural sources of airborne particulates in Europe are sea spray and soil resuspension by the wind. In addition, in the Mediterranean basin and the Atlantic archipelagos (eg Canaries, Azores), Saharan dust and volcano emissions can also be important natural sources of particles.

In Europe, ambient concentrations of PM₁₀ have been monitored in some urban networks since 1990, but there is currently no coherent overall European PM₁₀ data set, mainly because PM₁₀ has only been systematically monitored in a few member states. In addition, there is, as yet, no standardised method for monitoring PM₁₀ across Europe, although standardisation of PM₁₀ measurement methods is under development (cf. CEN/TC 264/WG6).

Previous studies and the data collected by the Working Group from Member States indicate that, though there does not appear to be a consistent pattern of concentrations between site types (urban background, traffic and industrial), the urban data do show a reasonably consistent pattern of lower concentrations in the far north of Europe and higher concentrations, possibly due to naturally occurring particles, in the southern countries.

Since PM₁₀ concentrations will be influenced by natural particles (which tend to be of larger size than man-made), the Group feel that in the future the measurement of the PM_{2.5} fraction and its standardization would enable a better comparison of anthropogenic particle concentrations throughout Europe to be made. At present, however, PM₁₀ as a PM metric seems to be a reasonable compromise between theoretical arguments favouring the measurement of very small particles and the knowledge and practical experience based on existing PM₁₀ and TSP (and BS) measurements.

2. RISK ASSESSMENT

There is increasing evidence that health effects occur at very low levels of PM, and without an apparent threshold. This evidence arises from studies, initially in the US, but which have more recently been carried out in Europe and elsewhere with similar conclusions. These studies have, in general, used different measures of particles, but the majority have used PM₁₀ and the Group felt that this was the most appropriate measure for a limit value for particles at the present time.

The health effect studies to date have not identified a clear no-effects threshold, and on this basis the extent of the adverse effects over a year are determined by the annual average concentration (multiplied by the appropriate regression coefficient). However, as the regression coefficient in the association of the adverse effects with particle concentrations is determined by the variation in 24-hour concentrations in the time series studies, the Group felt it appropriate to recommend both 24-hour and annual average limit values. It was also agreed that under practical conditions, absolute upper limits are not adequate. Therefore, an upper limit (24-hour) should be defined with few exceedances allowed, defined, for example, as a low number of accepted exceedances per year or as a 98 to 99.5 percentile. The final choice of the 98th percentile was influenced by its better stability.

The Group recognised the difficulties involved in setting limit values for particles when the epidemiological studies had not demonstrated a no-effects threshold. However, the Group felt that limit values could be recommended at levels at which public health effects at the population level were likely to be small. On this basis the Group initially considered ranges of daily values of 30-100µgm⁻³ and 15-40µgm⁻³ as an annual average. Drawing on studies in Europe and the US, and considering the WHO summary of exposure-response relationships the majority of the Group recommended a daily limit value of 50µgm⁻³ as a level at which public health effects were likely to be small. The Group recommended this limit value be adopted as a 98th percentile of daily values over a calendar year. The majority of the Group further recommended an annual average of 20µgm⁻³.

The Group puts forward these recommendations recognising that the project on economic evaluation has still to produce its final report, and that the Group's recommendation will be assessed in the light of the economic evaluation by the Steering Committee. Also, the question of geogenic dust, which plays an important role in some member states, has not been taken into full account and needs further consideration by the Commission. Whatever decisions the Commission takes finally on the above values, the Group recommends that they be reviewed within 5 years after entry into force.

Similarly, without an effects threshold, no alert values can be proposed on a scientifically sound basis. If, nonetheless, for specific reasons alert values are considered useful for some areas, local decisions on alert thresholds and adequate measures to be taken in case of an alert seem to be more useful than EU-wide regulations.

3. MEASUREMENT

The Directive requires both monitoring and assessment of air quality, and presentation of air quality data to the Commission.

3.1 MONITORING

Commercial manual and automatic instruments are available for monitoring TSP, PM₁₀ and other particle size fractions over the timescale appropriate for checking compliance with the limit value. Monitoring must be undertaken by a method shown to be equivalent to the nominated reference samplers or to one of the transfer reference samplers, and which gives data with the appropriate time resolution to check compliance with the limit value. The CEN standard methodology should be used to check samplers against the reference samplers.

In areas where monitoring is required, data capture over the year should be at least 75% for automatic methods and 90% of planned measurements for manual methods. Lower data capture may be acceptable in other areas.

Values for x and y (Article 6), defining zones for monitoring should be set at:

x: 70% for annual average 60% for high percentiles of daily averages

y: 50% “ 40% “

The number of monitoring stations per zone is defined as:

N_o in areas where assessment has been carried out

N_i in other areas

N_o is a minimum number of stations, i.e. 1

The Group recommend that values for N_i should be defined as follows:

Urban background sites per agglomeration	$2 \times \sqrt{\frac{I}{0.25}} = 4\sqrt{I}$
Roadside sites per country	$3 + \sqrt{P}$
Background sites per country	$\frac{A}{50000}$
Industrial sites per country	“sufficient to cover significant industrial sources”

Where:

I = the number of inhabitants of the agglomeration(millions)

P = the population of the country (millions)

A = the area of the country (square kilometres)

Where possible, laboratories undertaking monitoring should seek formal accreditation under EN45000 or Good Laboratory Practice, rather than more general quality standards such as EN ISO 9000. Quality control procedures should be fully documented and cover all aspects of the measurement process.

3.2 ASSESSMENT

Dispersion models used for assessments with regard to regulatory purposes need to be carefully validated against a reference situation to assure and control their quality. In addition to dispersion models, concentrations of PM_{10} can also be estimated using other pollutants as indicators, although this technique is not appropriate in locations where concentrations of PM_{10} and other pollutants are dominated by industrial pollutant sources with unpredictable or sporadic emissions. In order to be sure that decisions are properly founded, it is necessary to be certain that the recorded measurements or model calculations genuinely reflect the existing situation; in other words, the data must be of clearly defined and documented quality.

3.3 DATA PRESENTATION

Monitoring data showing levels, date, period and location of any exceedence of the limit value must be communicated to the Commission. It is suggested that a pro forma or protocol is established to achieve this in a harmonised way. In order to calculate percentile concentrations, it is recommended to use the method given in Directive 80/779/EEC (SO₂/SPM Directive). Full annual datasets should also be provided to the EEA.

4. COSTS

Patterns of particle emission are different from those of the other pollutants included in the first phase of the EU Framework Directive because of their extreme diversity of origin and source, both primary and secondary, natural and anthropogenic, and there are significant differences in levels between Member States and regions within states.

The limited amount of reliable PM₁₀ data makes it difficult to establish a comprehensive overview of PM₁₀ concentrations and trends in Member States. There is, therefore, a need to use detailed models which take dispersion, deposition and atmospheric chemistry into account. In turn, in order to assess the relative effectiveness and benefits of proposed abatement strategies and their likely health impact, it is important to consider the contributions of different source types to measured concentrations of particles using source-apportionment analysis or comprehensive emissions inventories, and dispersion models coupled to an exposure model.

The available data does, however, suggest that present PM₁₀ values exceed the recommended limit values in the majority of Member States. In future projections, planned actions should be taken into consideration. These include the SO_x and NO_x Protocols within UNECE and the Auto-Oil programme. Abatement policies for other pollutants will also have an impact. Actual reductions are, though, likely to be difficult to predict and not necessarily linearly related to emission reductions because of formation rates for secondary aerosols, which are in turn dependent on concentrations of other species in the air.

Thus, abatement strategies have to be developed if the currently planned precautions do not reduce emissions sufficiently, as seems likely. These strategies must include a definition of needs and a consideration of the risks, costs and benefits. In assessing costs, particularly in the context of abatement measures which control several pollutants at once, care must be taken not to assign the total costs to any one pollutant. On benefits, the primary consideration for the proposed limit values has been the protection of human health. As a threshold for zero risk could not be established, the eventual determination of limit values will involve the consideration of what constitutes acceptable risk. The health benefits should be evaluated by considering the outcome of the WHO Working Group, incorporating the more recent PEACE and APHEA studies. There are also likely to be a wide range of other benefits gained, though these are not yet adequately specified, including the limiting of buildings and materials damage and the soiling of plants.

In order to achieve the necessary reductions in particle levels, abatement strategies may have to take into account regional differences and be implemented at a variety of scales, given the variance and diversity in particle emissions outlined above. Furthermore, recommended limit values for particulate matter are based on PM₁₀ concentrations, which are only a fraction of the total aerosol in the atmosphere. A change to or inclusion of PM_{2.5} seems likely at some point in the future. It will thus be appropriate to consider to what extent PM_{2.5} levels will be influenced by proposed abatement measures for PM₁₀.

CONTENTS

1. INTRODUCTION AND POLLUTANT DESCRIPTION	7
1.1 BACKGROUND.....	7
1.2 DEFINITIONS.....	7
1.3 MAIN EUROPEAN PARTICULATE SOURCES	8
1.4 ANTHROPOGENIC SOURCES.....	8
1.4.1 Road Transport.....	8
1.4.2 Stationary Combustion Sources.....	9
1.4.2.1 Domestic and Service.....	9
1.4.2.2 Industrial	9
1.4.2.3 Urban and Industrial Incineration and Waste Disposal.....	9
1.4.2.4 Fossil Fuel Power Plants	9
1.4.3 Non Combustion Sources.....	10
1.4.3.1 Construction, Quarrying and Mining.....	10
1.4.3.2 Cement Plant and Ceramic Industry	10
1.4.4 Other Anthropogenic Sources.....	10
1.4.4.1 Forest and Agricultural Fires.....	10
1.4.4.2 Agriculture	11
1.4.5 PM ₁₀ Emissions in Europe.....	11
1.5 NATURAL SOURCES.....	11
1.5.1 Sea Spray.....	11
1.5.2 Soil Resuspension	11
1.5.3 Long Range Dust Transport.....	12
1.5.4 Volcano Emissions.....	12
1.6 CHEMISTRY	13
1.6.1 Formation of Aerosol Particles.....	13
1.7 DESCRIPTION OF CURRENT AMBIENT CONCENTRATIONS IN EUROPE.....	14
1.8 TRENDS IN EMISSIONS	16
1.9 SUMMARY OF KEY POINTS	16
2. RISK ASSESSMENT	18
2.1 RECOMMENDATIONS BY WHO.....	18
2.2 GENERAL CONSIDERATIONS.....	20
2.2.1 New Information.....	20
2.2.2 Implications of the shape of exposure-effect relationship.....	20
2.2.3 Effects of mixtures of air pollutants	22
2.2.4 Relevant health endpoints and exposure-effects relationships	22
2.2.5 Exposure-effects relationships.....	23
2.2.6 Results from APHEA and PEACE	25
2.2.7 Effect estimates based on the results of time-series analyses vs. cohort studies.....	26
2.3 NUMBER OF INDIVIDUALS CONCERNED IN EUROPE AT THE PRESENT TIME	27
2.4 DEGREE OF EXPOSURE AND IMPACT OF POLLUTANT	28
2.5 PRESENCE OF SENSITIVE SUBGROUPS.....	30
2.6 HIGHLIGHTING OF DIFFERENCES BETWEEN MEMBER STATES	30
2.7 RECOMMENDATIONS FOR LIMIT VALUES AND ALERT THRESHOLDS.....	31
2.7.1 An appropriate measure of particulate matter.....	31
2.7.2 Averaging periods.....	32
2.7.3 Alert Levels	32
2.7.4 Limit Values.....	32
2.8 RESEARCH NEEDS.....	34
2.9 SUMMARY OF KEY POINT AND RECOMMENDATIONS	35

3. MEASUREMENT	36
3.1 INTRODUCTION.....	36
3.2 MEASUREMENTS OF FINE PARTICULATES.....	37
3.2.1 <i>Introduction</i>	37
3.2.2 <i>Measurement methods</i>	37
3.2.2.1 Sample inlet.....	37
3.2.2.2 Collection medium.....	38
3.2.2.3 Mass Measurement.....	38
3.2.3.....	41
3.2.3 <i>Reference Sampler & Equivalence Reference Samplers</i>	41
3.2.4 <i>Number & frequency of measurements</i>	42
3.2.5 <i>Network Design</i>	43
3.2.5.1 Preliminary Assessment (Article 5).....	43
3.2.5.2 Monitoring Requirements (Article 6).....	43
3.2.5.3 Number of Monitoring Stations.....	44
3.2.5.4 Local Station Siting Requirements.....	46
3.2.6 <i>QA/QC Requirements</i>	47
3.2.6.1 Data Quality Objectives (DQOs).....	48
3.2.6.2 Monitoring Station Design.....	48
3.2.6.3 Equipment Evaluation and Selection.....	48
3.2.6.4 Site Infrastructure and Routine Operation.....	49
3.2.6.5 Maintenance and Calibration of Equipment.....	49
3.2.6.6 Data Review and Validation.....	50
3.3 ASSESSMENT OF FINE PARTICULATES.....	50
3.3.1 <i>Introduction</i>	50
3.3.2 <i>Assessment requirements</i>	50
3.3.3 <i>Assessment models for particulates</i>	51
3.3.3.1 Long term (yearly average).....	51
3.3.3.2 Short term (percentiles).....	51
3.3.4 <i>Other Assessment Techniques</i>	51
3.3.5 <i>QA/QC for assessments</i>	52
3.4 DATA PRESENTATION AND EVALUATION.....	52
3.4.1 <i>Introduction</i>	52
3.4.2 <i>Presentation of Measurements</i>	52
3.5 SUMMARY OF KEY POINTS AND RECOMMENDATIONS.....	53
3.5.1 <i>Measurement</i>	53
3.5.2 <i>Assessment</i>	54
3.5.3 <i>Data Presentation</i>	55
4. COSTS	56
4.1 INTRODUCTION.....	56
4.2 ABATEMENT TECHNIQUES.....	57
4.3 BENEFITS OF THE REDUCTION OF PM ₁₀	58
4.4 DISCUSSION.....	59
5. REFERENCES	60
6. ANNEX 1: PARTICULATE MATTER EMISSIONS INVENTORIES	65
7. ANNEX 2: PARTICLE CONCENTRATION DATA FROM MEMBER STATES	68
8. ANNEX 3: UNCED AGENDA 21 AND AIR QUALITY MONITORING	85

1. INTRODUCTION AND POLLUTANT DESCRIPTION

1.1 BACKGROUND

Airborne suspended particulate matter can be of primary origin, i.e. emitted directly into the atmosphere or of secondary origin, i.e. formed in the atmosphere from gaseous species by either homogeneous or heterogeneous chemical reactions. Due to these different emission sources, particles have different chemical composition and size distributions. Depending on their size, particles have a different potential to be transported over either long or short distances [1].

Primary particles can be produced from either natural or anthropogenic sources. In Europe the main natural sources are sea spray [2] and soil resuspension by wind [3]. Also, in the Mediterranean basin, Saharan dust [4] and volcano emissions [5] are important contributors to airborne particulates. On the other hand, the sources of anthropogenic emissions of particles are similar across Europe. The most common sources are related to traffic, power plants, combustion sources (industrial and residential), industrial fugitive dust, loading/unloading of bulk goods, mining activities, man-made forest fires and in some local cases non-combustion sources such as building construction and quarrying.

Secondary particles can also have a natural or anthropogenic origin. Natural particulate sulphate is formed by oxidation of dimethyl sulphide produced by sea phytoplankton. Also, gaseous ammonia can be naturally emitted and can produce secondary particles by reacting with acidic gases [6]. In addition, in the Mediterranean basin, sulphur dioxide emissions from volcanoes which can react to form secondary particles, can be substantial [7]. However, in most European countries industrialisation and high volumes of traffic mean that anthropogenic sources should be predominant, particularly in urban areas.

1.2 DEFINITIONS

For the purpose of this position paper the following definitions are applied:

Ambient aerosol concentration: The mass concentration of aerosol particles present in the air before the particles are affected by the presence of the sampler.

Accumulation mode: A part of the size spectrum of airborne particles, between approximately 0.1-2 μm diameter, in which particles have a long atmospheric lifetime.

Nucleation mode: A part of the size spectrum of airborne particles, below about 100 nm diameter, in which particles arise mostly from fresh emissions from combustion processes, and gas to particle conversion.

Suspended Particle Matter (SPM): SPM is the notion of all particles surrounded by air in a given, undisturbed volume of air.

Black Smoke (BS): Strongly light-absorbing particulate material suspended in the ambient atmosphere. By convention measured by light reflectance of a filter stain [8,9].

Total Suspended Particulates (TSP): By convention the estimate of SPM concentration as provided by the classical US - High Volume Samplers.

Fine particulate matter: For the purposes of this position paper, fine particulate matter is defined as specific fractions of suspended particulate matter as follows:

PM₁₀: A target specification for sampling instruments as defined in the US Federal Register [10] to mimic the thoracic fraction. It represents particles with an aerodynamic diameter of 10 µm or less, or, more strictly, particles which pass through a size selective inlet with a 50% efficiency cut-off at 10 µm aerodynamic diameter.

PM_{2.5}: Particles with an aerodynamic diameter of 2.5 µm or less, or, more strictly, particles which pass through a size selective inlet with a 50% efficiency cut-off at 2.5 µm aerodynamic diameter.

Thoracic Fraction: The mass fraction of inhaled particles which penetrate beyond the larynx [11].

1.3 MAIN EUROPEAN PARTICULATE SOURCES

Examples of national emission inventories for the UK, Germany and the Netherlands are given in Annex 1. It is important to recognise that there are significant uncertainties in estimates of emissions of particles, so that detailed comparisons of the estimates in Annex 1 are not warranted. Moreover, although the estimates of emissions in the UK and Netherlands are for PM₁₀, those for Germany are for total particulate matter. While the great majority of the combustion emissions will be in the PM₁₀ category, this is not likely to be the case for the other sources.

It is also important to recognise that inventories for specific urban areas may differ significantly from national total emissions, as illustrated by the emission inventory for London (Table A 1.4); also, that the importance of sources contributing to certain episodes may differ from the picture presented in the annual inventories. In particular, emission inventories for particles in general contain no information on sources of secondary particles, which in many areas can form a significant proportion of ambient concentrations.

1.4 ANTHROPOGENIC SOURCES

1.4.1 Road Transport

Particulate emissions from road transport arise as direct emissions from vehicle exhausts, tyre and brake wear and resuspension of road dust. In urban areas, emissions from road transport are thought to be the major source of PM₁₀. In general, diesel engine vehicles emit a greater mass of fine particulate matter, per vehicle, than petrol engines.

Diesel emissions are mainly composed of soot particles, volatile hydrocarbons and some sulphate from the fuel sulphur. When hydrocarbons and sulphates are released by the car exhaust they condense on airborne particles, mainly on the freshly emitted carbon. The size distribution of these particles tends to be bimodal, with particles of 0.01 to 0.05 µm in the nucleation mode in the case of freshly emitted soot particles and, of some 0.05 to 2.5 µm in the accumulation mode in the case of older coagulated soot particles.

The movement of vehicles on the street also results in resuspension of road dust. Emissions also occur as a result of tyre wear and brake lining wear. Although there is a lack of data, it is expected that most of these particles will be in the size range 3 to some 30 µm. The chemical composition of these particles may also be very different from those derived from combustion.

The road dust deposit available for resuspension comes from mechanical wear of, and dirt on, vehicles (incl. tyre and brake lining wear), debris from loads on vehicles, influx of soil material etc. In some parts of Europe, notably in Scandinavia, the widespread use of studded tyres on vehicles wears off the road surface (asphalt, concrete) to such an extent that the resulting particle deposit on the road becomes the

totally dominating source of resuspension particles. This source is active during dry winter situations on snow/ice-free roads and causes substantially increased PM₁₀ concentrations.

1.4.2 Stationary Combustion Sources

1.4.2.1 Domestic and Service

Domestic coal burning was traditionally a major source of particles in European cities during winter months. Nowadays, however, abatement strategies mean that this source has decreased in significance. Only in some northern European cities, such as Belfast [12-14], where coal is still used as a domestic fuel, is domestic coal burning a significant particle source.

1.4.2.2 Industrial

Industrial emissions can be a significant source of particulate emissions in urban areas. The contribution that this source makes to ambient particulate material will vary depending on the location of the industry and the abatement technology adopted. Although many studies have been conducted to characterise emissions from large industrial sources, for example steel works, information on small urban emitters, for example metallurgical processes and small factories, is more limited. Particles emitted from industrial sources have been found to be in the size range 0.5 to some 100µm, depending on the nature of the source. Composition will also depend on the nature of the source.

1.4.2.3 Urban and Industrial Incineration and Waste Disposal

In many European countries incineration is used as an important means of waste disposal in urban areas. The proportion of waste disposal by this route varies across Europe, from 13% in Italy to 53% in Switzerland, with an average value of 20 %. However, in some countries (for example, Portugal, Spain) the use of incineration is not used or is under consideration as a new option for waste disposal.

Two main types of pollutants (combustion gases and fly ash) are emitted from incinerators. Fly ash is composed of soot, trace metals, mineral dust, and partially burnt material with a size distribution between 5 and some 150µm. Both the size of the fly ash and the amount emitted are specific to individual incinerators and on the nature of the particulate scrubbers in use. Also, for a specific incinerator the particle emission varies during operation. Finally, ash recovered in the scrubbers system has to be correctly disposed of, in a controlled landfill, to avoid the emission of fugitive dust [15].

Accidental fires originating in uncontrolled waste landfill can produce high levels of particulate emission. However, this source is episodic and will only have a local impact.

1.4.2.4 Fossil Fuel Power Plants

Coal fired power stations usually have abatement equipment such as cyclones or electrostatic filters which remove, on average, 99.5% of particulate emissions. Therefore, only the fine fraction, which can be transported by wind, is expected to contribute to an increase in ambient levels of suspended particulate matter in other areas. In contrast, oil fired power stations are not equipped with precipitators in some member states but, even so, generally emit less particulate matter per MWh generated than coal fired power stations. Some countries in Europe are increasingly utilising natural gas to generate electricity. Particulate emissions from this type of power station are even smaller.

The CORINAIR study [16] showed power stations as the main stationary source of sulphur dioxide in Europe. Part of this SO₂ will undergo chemical reactions producing sulphuric acid which leads to the formation of secondary particles. In the case of very high chimneys, most of the emitted fine particles and the secondary acidic aerosol can be transported over continental distances.

1.4.3 Non Combustion Sources

1.4.3.1 Construction, Quarrying and Mining

Although data quantifying particulate emissions from construction and demolition work is scarce, some research carried out in USA gives an estimation for TSP of 2.5 tonnes/hectare/month in zones where large construction work is in progress. The quantity of particles emitted in each city from this source will depend on the type of construction in progress. These particles are mainly present in size fractions greater than 10µm. However, some fraction of the total amount is likely to be present as smaller particles. Also, some of this dust will be resuspended either by traffic or wind.

It is also difficult to assess the amount and composition of fugitive dust emitted from quarrying and mining activities. US emission factors for mineral handling, quarrying and mining range from 0.007 to 0.119 kg/tonne of waste produced [75]. Depending upon the mechanical activity, rock type and wind speed, the majority of the mass of this fugitive aerosol is expected to be present in general in sizes above 3µm particle diameter. A small amount of these particles can also be contained in the size range between 1 and 3 µm.

1.4.3.2 Cement Plant and Ceramic Industry

It is difficult to assess the percentage of airborne particles emitted either from cement plants or from ceramic industries in urban aerosol, due to the similarity of their chemical composition to soil or construction dust. In the UK, cement and lime manufacture has been estimated to produce 4456 tonnes of total particulate matter a year.

1.4.4 Other Anthropogenic Sources

1.4.4.1 Forest and Agricultural Fires

Both direct emission from fires and ash resuspension from burnt soils could be an important source of airborne PM₁₀. This pyrogenic material, which is composed of organic matter, black carbon and inorganic material, is to a large extent present in the size range below 10 µm and so can be resuspended by wind [17].

Although the contribution to ambient aerosol from fire smoke will generally be episodic, in areas where there is a constant forest burning, the particulate emission from this source could be significant. Each year in Mediterranean countries about 50 000 fires are set (mostly by man) producing between 700 000 to 1 000 000 of hectares of burnt land [18]. This process can therefore make an important contribution to airborne particulate matter in the aforementioned countries.

Agricultural fire emissions are mainly due to stubble burning. However, this source is episodic and has been banned in some European countries. It is therefore not expected to be a significant source of

particles. Other fires may be essential to the maintenance of certain types of habitat such as heathland and heath moorland.

1.4.4.2 Agriculture

Emissions of wind-blown soil dust can occur from bare fields, especially in dry periods. These particles are likely to be relatively large and will not, in general, contribute significantly to overall PM_{10} levels. The most important source of fine particles from agricultural activities results from the reaction of ammonia (of which agriculture is the dominant source in most of Europe) with sulphuric and nitric acids which are the products of fossil fuel combustion. The resulting ammonium sulphate and nitrate aerosols can form a major component of ambient PM_{10} levels in many areas.

1.4.5 PM_{10} Emissions in Europe

At present it is difficult to give a comprehensive overview of the PM_{10} emissions in the European countries. One of the problems is that most of the available information refers to particulate matter in general and is not specific to PM_{10} . In addition not all countries have emission inventories that are detailed enough to specify all kinds of sources. Annex 1 gives information on PM_{10} emissions and major sources for three European countries. It must be emphasised that the numbers given in these tables do not contain secondary aerosols and that the emission sources listed are not comparable in all cases.

1.5 NATURAL SOURCES

1.5.1 Sea Spray

Breaking waves on the sea cause the ejection of many tiny droplets of seawater into the atmosphere. These droplets dry by evaporation leaving sea salt particles suspended in the air. Particles are also directly emitted by the bursting of air bubbles on sea surface. Such particles are generally in the size range between 1 to 20 μm . [19]. Whilst these particles are, in the main, rather coarse in size, a minor part of their mass is in particles small enough to have an appreciable atmospheric lifetime, which has been estimated as three days [20]. Clearly, coastal areas will be the most affected, but sea salt is also measurable at inland locations.

Airborne sea salt shows a similar chemical composition to sea salt, with anions (chloride and sulphate), cations (sodium and magnesium) and organic phosphorus. Also, trace metals (cadmium, lead, vanadium, and zinc) have been found in marine aerosol. This aerosol metal enrichment arises from bubbles of water scavenging before bursting.

In winter months, identification of this source by measurement of the chemical composition of airborne particles is complicated by the use of salt for de-icing the roads. Most road de-icing salt has a chemical composition almost identical to that of sea salt and the two are effectively indistinguishable. Vehicles travelling at high speed on the motorway in wet conditions raise very visible plumes of spray which contain salt if the road has been treated with de-icing salt. Although the process is less visible, it occurs also on urban roads at lower traffic speeds.

1.5.2 Soil Resuspension

Meteorological mechanisms such as wind, temperature changes and water produce soil dust by either rock or mineral weathering. This dust can be carried by wind and has a particle size distribution depending

upon its original geological source [21] and can be in the size range between 5 to 50 μm . However, fine sand has a log normal distribution around a particle size of about 10 μm . The chemical composition of soil particles is similar to their geological origin as dolomite, gypsum quartz and clay minerals. Usually, an analysis showing enrichment in silicon, calcium, iron and aluminium in the aerosol indicates its geological origin.

The action of the wind on dry loose soil surfaces leads to particles blowing into the air. Factors favouring the suspension of soil dust particles into the atmosphere are an exposed dry surface of fine soil and a high wind speed. In towns and cities, the areas of exposed soil, particularly in town centres, are rather small. However, there are considerable quantities of dusts on road and pavement surfaces which arise from ingress of soil on vehicle tyres and from the atmosphere, the erosion of the road surface itself and degradation of parts of the vehicle, especially the tyres. Because these particles lie on a surface which readily dries and is subject to atmospheric turbulence induced by passing vehicles, this provides a ready source of particles for resuspension into the atmosphere. The amounts of dust resuspended in this process are extremely difficult to predict or measure, as they depend critically upon factors such as the dust loading of the surface, the preceding dry period and the speed of moving traffic. However, the size distribution and chemical composition of particles in the urban atmosphere give a clear indication that this source can contribute significantly to the airborne particle loading of our cities.

1.5.3 Long Range Dust Transport

A source of airborne particulate matter that cannot be neglected is the injection of windblown natural dust into the atmosphere in sand and dust storms common during windy conditions in the world's deserts. In the northwestern Mediterranean region, the input of Saharan material, known locally as red rains, has been estimated as 3.9 million of tonnes each year [22]. In some parts of the Mediterranean basin, it is thought that this makes a substantial contribution to local airborne particulate matter. Although the main concentration of Saharan dust is in the Mediterranean area, Saharan dust is also transported to Northern Europe [20], and other continents such as America [23].

These processes do extend to other regions of the globe, although their magnitude is obviously reduced where soils are moist and have vegetation cover. Much enhanced deposition is occasionally seen when the atmosphere carries dust from the Sahara desert regions. Such particles are generally rather coarse (i.e. large in size) which usually have only a limited atmospheric lifetime and range but their transport from North Africa to Northern European countries shows that this is not always the case. The composition of Saharan dust shows a high content in calcite with some amount of gypsum, clay minerals and metals which affects the atmospheric chemistry in the Mediterranean countries [24]. However, direct assessment of the percentage contribution of Saharan dust to PM_{10} in the Mediterranean area needs further research.

1.5.4 Volcano Emissions

Volcanic activity still occurs on some islands off continental Europe, such as the Canaries, Iceland and Sicily. Fine fly ash emitted from such volcanoes could represent an important local source of PM_{10} particles in nearby cities. Emissions of sulphur dioxide (SO_2) from volcanoes can also contribute to the formation of secondary particles. For instance, Mount Etna provides a semi-permanent emission of SO_2 (4,000 tonnes/day) from volcanic plume and magma degassing [5].

1.6 CHEMISTRY

1.6.1 Formation of Aerosol Particles

After its emission or secondary formation, the length of time airborne matter will remain suspended in the air will depend upon its density, shape and size and meteorological conditions. Suspended particles are deposited by dry deposition, either by sedimentation and gravitational settling or impaction due to atmospheric turbulence and diffusion. This latter process is characteristic of particles which undergo Brownian movement and sizes below $0.1\mu\text{m}$. While dry deposition accounts for 25% of particle atmospheric removal, wet deposition by rainout or washout is more effective at removing particles from the atmosphere.

The mechanism of formation is crucial in influencing the size of particulate aerosol. Atmospheric aerosol shows evidence of several categories of sources in its size distribution. Typically three major components are recognised [25]. The first one, called nucleation mode, contains very large numbers of particles with a diameter of about $0.1\mu\text{m}$ (100nm). These fine particles can be formed as a result of combustion processes and evaporation from hot surfaces or from gas phase reactions in the free atmosphere to form involatile molecules. Nucleation particles have a short lifetime of a few hours because they coagulate with larger particles or act as condensation nuclei.

Particles with diameter between about 0.05 to $2.5\mu\text{m}$ contained in the so-called accumulation mode, are mainly formed by coagulation and generated from condensation processes. Particles in this size range can also be produced from secondary formation and natural sources such as sea spray or the resuspension of fine soil or dust, although less than 10% of mass concentration of the fine fraction ($<2.5\mu\text{m}$) has a mineral origin. Therefore, the measurement of $\text{PM}_{2.5}$ should improve the comparison of particle concentrations between countries with low natural emissions (Northern Europe) and others which undergo high natural particle input (Southern Europe). Particles in this size range are long lived in the atmosphere, since their removal mechanisms are least efficient in this region, and are therefore important vectors for long-range transport.

Secondary aerosol is produced by the oxidation of primary gases (sulphur dioxide, SO_2 , nitrogen oxides, NO_x , and volatile organic compounds, VOCs,) to sulphuric and nitric acid, and organic vapours, followed by their gas-particle conversion [26, 77]. Finally, some of these acidic gases can be neutralised by reaction with ammonia gas or calcium carbonate (calcite) forming secondary aerosol (ammonium sulphate, ammonium nitrate, calcium sulphate, calcium nitrate). In urban agglomerations the latter process will be predominant, due to the high anthropogenic emission of SO_2 and NO_x from domestic heating and traffic emissions. However, emissions of natural SO_2 and VOCs (terpenes) could play an important role in the formation of secondary aerosol. The photochemical nature of the process that produces organic matter from VOCs implies that their main contributions to PM_{10} concentrations will be found during summer.

Both nucleation and accumulation modes are commonly defined as fine particle modes while particles with diameters above $2.5\mu\text{m}$ are contained in the so-called coarse mode, with a peak of some $10\mu\text{m}$. This coarse fraction is the third major component of particles found in air. These are shorter lived and are likely to travel distances of typically metres to hundreds of kilometres, according to size and wind speed. They may contribute substantially to aerosol mass, although the numbers of such particles is often small.

Coarse particles can be formed by rock weathering producing particles with a high content of minerals, sea spray and industrial processes. Coarse particles have been found to consist of 50% of geological material [27].

These three components overlap to comprise the broad size distribution observed in the atmosphere [28, 29]. The greatest number of particles is almost invariably in the region smaller than $0.1\mu\text{m}$ diameter. The mass of the particles, however, is predominantly in the accumulation and coarse particle fraction.

Airborne particle size is also relevant to the particulate inhalation properties. Particles with a size well above $2.5\mu\text{m}$ can be deposited in the nose and larger particles in the size range above $10\mu\text{m}$ deposited in other airways of the head. On the other hand, particles of below $10\mu\text{m}$ (thoracic fraction), can pass beyond the larynx and enter the human thoracic airways. However, only fine particles below about $6\mu\text{m}$ are deposited in the lung alveoli [30]. A further description of penetration and deposition of particles in the respiratory system is given in reference 38.

1.7 DESCRIPTION OF CURRENT AMBIENT CONCENTRATIONS IN EUROPE

In Europe, ambient levels of PM_{10} have been monitored in some urban networks since 1990. However, there is no coherent European PM_{10} data set since there is no standardised method for monitoring PM_{10} across Europe. In addition, there are only a few cities across Europe where monitoring has been carried out for a significant length of time using continuous instruments.

The working group has attempted to collect together information from Member States on monitoring of ambient levels of PM_{10} across Europe from sites operated continuously for a significant amount of time. These results are given in Annex 2. In addition to PM_{10} data, TSP data were supplied by Italy and Finland and Black Smoke data by France, the UK and Ireland. These data are also included in Annex 2, but are not further discussed.

Analysis of these European PM_{10} data shows that levels vary both across Europe and within individual countries. Annual mean PM_{10} concentrations vary from $10\mu\text{g m}^{-3}$ in remote regions [31] to above $100\mu\text{g m}^{-3}$ in urban industrial areas in some countries.

The data illustrate that, within each country, annual mean PM_{10} concentrations remained reasonably constant during the three year period with an interyear variability of less than 50 % (cf. section 3.2.5.2). The same general consistency is found in the analysis of 98th percentile of daily values. Hence, for further analysis of these data, the results of data for all three years are used together.

Table 1.1: Summary of PM₁₀ Concentrations at Urban, Traffic and Industrial Sites in Europe 1992 - 1994

Country	Number of Sites	Annual Mean $\mu\text{g m}^{-3}$	98th Percentile of Daily Means $\mu\text{g m}^{-3}$
Urban Background (UB) Sites			
United Kingdom	13	20 - 34	41 - 95
Luxembourg			
France	3	41 - 67	68 - 136
Netherlands	4	37 - 41	92 - 126
Belgium			
Portugal	1	72 - 75	144 - 146 (95th%ile)
Spain	5	39 - 89	85 - 222
Sweden	5	12 - 16	
Finland	3	22 - 25	
Germany	1	42 - 43	95
Urban Traffic (UT) Sites			
United Kingdom			
Luxembourg	1	30	61
France	2	51 - 54	94 - 136
Netherlands	4	39 - 43	90 - 129
Belgium			
Portugal			
Spain			
Sweden	1	35	
Finland	10	13 - 28	43 - 89
Germany	2	36 - 65	77 - 98
Urban Industrial (UI) Sites			
United Kingdom			
Luxembourg	1	32	71
France	9	43 - 78	58 - 143
Netherlands			
Belgium			
Portugal			
Spain	4	52 - 123	106 - 291
Sweden			
Finland	5	14 - 19	32 - 56
Germany	1	50 - 58	128

UB = Urban Background: a site in a central urban area not influenced by a single major source (including a road), and not necessarily mainly residential.

UT = Urban Traffic: a site in a central urban area influenced by and close to a major road.

UI = Urban Industrial: A site in a central urban area influenced by industrial sources.

Table 1.1 summarises concentrations found at different types of urban sites. The table contains data from Annex 2 that have been classed as urban background (UB), urban traffic (UT) or urban industrial (UI) by the countries. It shows that there does not appear to be a consistent pattern of concentrations between these site types, as might be expected. This is probably because of the low numbers of sites for which data are available, especially for traffic sites. However, the data do show a reasonably consistent pattern of lower concentrations in the far north of Europe and higher concentrations in the southern countries. (Annual average concentration range: Sweden 12-16 μgm^{-3} , UK 20-34 μgm^{-3} , Netherlands 37-41 μgm^{-3} , Germany 42-43 μgm^{-3} , France 41-67 μgm^{-3} and Spain 39-89 μgm^{-3} .)

A better picture of PM_{10} concentrations within and between the countries of Europe will only emerge as the number of monitoring sites increases and harmonised monitoring and QA/QC procedures are adopted. This will arise primarily as a result of the implementation of the EC Daughter Directive.

1.8 TRENDS IN EMISSIONS

The emission inventories for the UK, Germany and the Netherlands suggest that there has been a decline in particulate matter emissions over time, although the picture varies from country to country and the period covered is often short (Annex 1). These national figures can, though, hide the fact that the relative importance of different sources can vary significantly from one area to another. For instance, in the 1990 inventory for the UK, road transport accounted for 25 % of PM_{10} emissions, while in London road transport accounted for 85 % of emissions (Annex 1). Similarly, the relative importance of sources during episodes can vary from the annual picture. For instance, studies in the UK have shown that during winter episodes road traffic may contribute some 75-85% of atmospheric concentrations of PM_{10} [14].

Due to already planned reductions in diesel engine emissions and industrial combustion emissions and the continuing decrease in domestic coal use, some reduction is anticipated in future years. Results from the Auto-Oil programme suggest that under a business-as-usual scenario European average exhaust emission of particles from urban diesel transport could decline to about 70 % of the 1990 level by the year 2010. In several countries, including Spain and Greece, these urban emissions are projected to be higher than the 1990 levels [73]. As a result of the package of Auto-Oil measures proposed by the European Commission urban particulate emissions from diesel transport in the EU are expected to fall to a level of 34 % of the 1990 level by the year 2010 [74]. It is important to note, however, that this is an average figure and the actual percentage will vary between EU countries and cities.

1.9 SUMMARY OF KEY POINTS

Airborne suspended particulate matter can be either primary or secondary in nature. Primary particles are emitted directly into the atmosphere whereas secondary particles are formed in the atmosphere from gaseous species.

Primary particles can be emitted by either natural or anthropogenic processes.

In most European countries, industrialisation and high volumes of traffic mean that anthropogenic sources predominate, especially in urban areas.

Emission sources of anthropogenic particles are similar throughout Europe. The most significant of these are traffic, power plants, combustion sources (industrial and residential), industrial fugitive dust, loading/unloading of bulk goods, mining activities, human-started forest fires and, in some local cases non-combustion sources such as building construction and quarrying.

The main natural sources of airborne particulates, in Europe, are sea spray and soil resuspension by the wind. In addition, in the Mediterranean basin, Saharan dust and Volcano emissions can also be important natural sources.

Secondary particles are formed from the oxidation of sulphur dioxide, nitrogen oxides and VOCs in the atmosphere and they have a predominantly man made origin.

In Europe, ambient concentrations of PM_{10} have been monitored in some urban networks since 1990. Currently, there is no coherent European PM_{10} data set, mainly because PM_{10} has only been systematically monitored in a few member states. In addition, there is no standardised method up to now for monitoring PM_{10} across Europe. However, standardisation of PM_{10} measurement methods is in progress (cf. CEN/TC 264/WG6).

The data collected by the Working Group from Member States indicate that, though there does not appear to be a consistent pattern of concentrations between site types (urban background, traffic and industrial), the urban data do show a reasonably consistent pattern of lower concentrations in the far north of Europe and higher concentrations in the southern countries, possibly due to a contribution from naturally occurring particles. PM_{10} measurements performed during the PEACE study (cf. 2.2.1) showed a similar pattern [39], although only preliminary results from this study have been considered.

In the future the measurement of the $PM_{2.5}$ fraction and its standardization would produce more reliable data in order to obtain a better comparison of anthropogenic particle concentrations through Europe.

2. RISK ASSESSMENT

This section of the report relies heavily on the assessment of the health effects of particles undertaken over the past two years or so by the World Health Organisation (WHO). As such, there is no explicit quantified consideration of effects on, for example, ecology or the built environment. This is due to the lack of quantitative information available. In its initial assessment in 1994, the WHO primarily considered evidence from the US with few European studies. Since that time more European work has been carried out, notably in the APHEA and PEACE studies (see below) and these have been incorporated into the WHO assessment.

2.1 RECOMMENDATIONS BY WHO

In the framework of the update and revision of the WHO Air Quality Guidelines for Europe, a Working Group on "Classical" Air Pollutants including particulate matter met in Bilthoven in October 1994 [32]. After a Final Consultation in October 1996, a final text is being prepared to be published in 1997. The currently available draft of 31 October 1996 [33] states that:

- short-term variations in PM exposure are associated with health effects even at very low levels of exposure;
- the current database does not allow the derivation of a threshold below which no effects occur;
- epidemiological studies are unable to define such a threshold, if it exists, precisely;
- at low levels of (24h) exposure (0 to 100 $\mu\text{g m}^{-3}$ PM₁₀), the exposure-response curve probably fits a straight line reasonably well;
- no guidelines for short-term or long-term average concentrations are recommended by WHO; risk managers are referred to risk estimates provided in tables for guidance in decision making regarding standards for PM to be set.

Table 2.1 gives the most important of the risk estimate tables. It is a summary of short-term exposure-response relationships for several health endpoints. Table 2.2 shows a conversion of the Relative Risks given in Table 2.1 to estimated relative changes in health outcome (Δ HI) associated with changes of daily average concentrations of PM₁₀ (Δ C).

Table 2.1: Summary of Relative Risk estimates for some health endpoints and PEF changes associated with a $10 \mu\text{g m}^{-3}$ increase in the concentration of PM_{10} or $\text{PM}_{2.5}$, according to WHO Working Paper [33]. LRS = Lower Respiratory Symptoms, PEF = Peak Expiratory Flow, n.a. = not available

	PM2.5	PM10
Health Endpoint	Relative Risk (95% C.I.)	Relative Risk (95% C.I.)
Bronchodilator use	n.a.	1.0337 (1.0205-1.0470)
Cough	n.a.	1.0455 (1.0227-1.0687)
LRS	n.a.	1.0345 (1.0184-1.0508)
Respiratory hospital admissions	n.a.	1.0084 (1.0050-1.0117)
Daily mortality	1.0151 (1.0112-1.0190)	1.0070 (1.0059-1.0082)
	Effects on PEF	Effects on PEF
PEF change (relative to mean)	n.a.	- 0.13% (-0.17%, -0.09%)

Table 2.2: Summary of short-term exposure-response relationships for some health endpoints (for PM_{10})

Health effect indicator(HI)	$\Delta C (\mu\text{g}/\text{m}^3)$	$\Delta \text{HI} (\%)$	$\Delta C (\mu\text{g}/\text{m}^3)$	$\Delta \text{HI} (\%)$
Daily mortality	71	5	10	0.7
Hospital admissions for respiratory conditions	60	5	10	0.8
Prevalence of cough in general population	15	5	10	3.4
Symptom exacerbations among asthmatics	11	5	10	4.6
Peak expiratory flow (mean change in popul.)	385	5	10	0.13

2.2 GENERAL CONSIDERATIONS

2.2.1 *New Information*

New information published since the WHO Working Group meeting in October 1994 has been considered in the update procedure of the WHO Air Quality Guidelines. This includes the two co-ordinated air pollution epidemiology studies conducted in Europe with support by CEC, the first results of which are now available.

The APHEA (short-term effects of Air Pollution on Health: a European Approach) project (co-ordinator Klea Katsouyanni, Athens, Greece) is an attempt to provide quantitative estimates of the short-term health effects of air pollution based on time series analyses in single locations, and subsequent meta-analyses. Health outcome data are daily counts of total and cause-specific deaths and hospital admissions. Exposure data consist of daily measurements of black smoke, sulphur dioxide, suspended particles, nitrogen dioxide, and ozone. Data have been collected from 15 cities with a total population exceeding 25 million in 10 European countries [39, 34, 36].

The PEACE (Pollution Effects on Asthmatic Children in Europe) study also uses time series analyses. It is a panel study conducted by 14 research centres in 10 countries (co-ordinator Bert Brunekreef, Wageningen, NL). Health outcome data are peakflow, respiratory symptoms and medication use in schoolchildren with chronic respiratory symptoms, exposure data consist of daily measurements of PM₁₀, black smoke, nitrogen dioxide, and sulphur dioxide [39].

2.2.2 *Implications of the shape of exposure-effect relationship*

The assumption of linearity in exposure-effect relationships is a simplification which seems to be adequate for a small range of concentrations frequently found during study periods. However, such an assumption may lead to erroneous assessments at concentrations representing the high or the low end of the concentration distribution during study periods or under real exposure conditions. At concentrations amounting to a few hundred $\mu\text{g m}^{-3}$ (as seen in, e.g., Erfurt, former GDR [44]), the slope of the exposure-effect curve has been shown to decrease, but this range would be mainly of interest with respect to alert levels.

The lower end of the concentration range is much more important but unfortunately not subject to direct exploration by experimental or epidemiological methods due to the substantial influence of causes other than exposure to low concentrations of air pollutants. For an individual, a threshold for health effects of pollutants may exist, which in turn may vary with the actual health state. In a large group of individuals or in a population, however, one can always assume that some individuals exist who are more susceptible than the most susceptible individual identified so far. The potential number of individuals reacting at very low concentrations will be smaller with lower concentrations. This response-frequency curve (x-axis: susceptibility, y-axis: frequency of reacting persons in a population) is assumed to be bell-shaped like many other biological distributions [cf. 72]. A bell-shaped response-frequency curve, however, necessarily results in an exposure-effect relationship which is S-shaped, i.e., which shows a slope decreasing with approximation both to low concentrations and to high concentrations (Figure 2.1). Under these circumstances, a clear threshold below which absolutely no (exposure-related) health effects occur is not very probable. Nevertheless, there may be a range of low concentrations associated with health effects considerably smaller than those that would be predicted in case of linearity of the exposure-effect relation down to the lowest concentrations.

The situation is illustrated in Figure 2.1. Here, the y-axis gives the “Total Health Outcome” (THO), i.e., the health outcome due to all causes (not just the part of the health outcome due to the exposure under

consideration). In the case of mortality this can be the total number of deaths per day in a defined population. Certain numbers of these deaths are associated with ambient air pollution, but the majority are due to other causes.

If exposure increases from I to K, THO will increase from D to C (Solid line). In case of linearity down to zero exposure (asterisk line), THO without any exposure (i.e., that one due to other causes) would amount to G, and an increase of exposure from zero to H would result in a THO of F which consists of the basal health outcome G plus an exposure-related effect of F minus G. In the case of an S-shaped relationship (dashed line), the basal health outcome at zero exposure would be F, and an increase of exposure from zero to H would result in a THO of E which consists of the basal health outcome F plus the exposure-related effect of E minus F. Thus, both the estimate of the basal health outcome without exposure (i.e., that one associated with causes other than exposure) and the estimate of the effect of relatively low exposures are greatly influenced by the shape of the exposure-effect relationship at low levels of exposure. Unfortunately, as mentioned above, a decision which shape of the curve is the “real” one cannot be based on direct measurement.

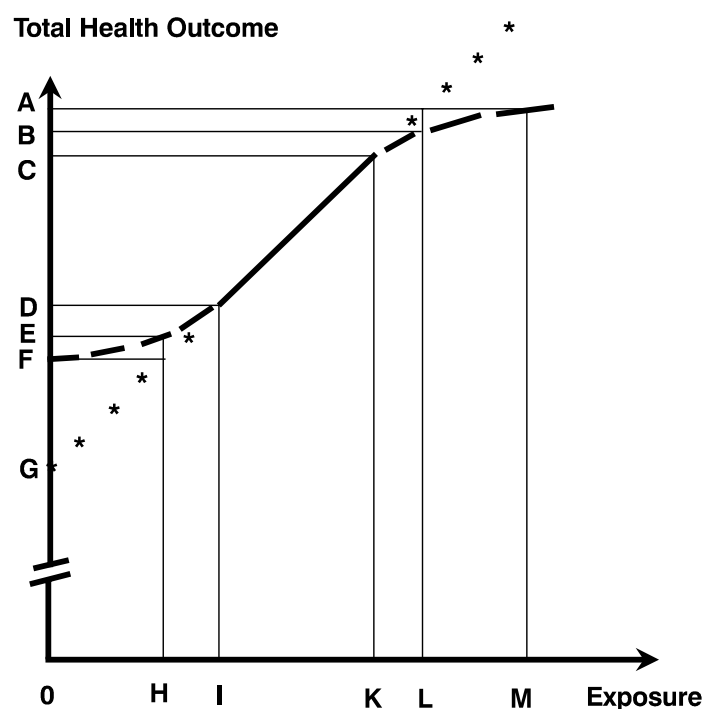


Figure 2.1: Hypothetical exposure-effects relationship. Implications of non-linearity. Note: Total Health Outcome means health outcome due to all causes, not only the part associated with exposure.

This discussion may seem to be very academic. However, the shape of the exposure-effects relationship and the presence or absence of a threshold are of considerable importance in health risk assessment. If a threshold exists which is exceeded only during a few days per year, adverse health effects are assumed to only occur at these few days. Without a threshold, however, due to the large number of days with average or relatively low concentrations, most of the health effects attributable to air pollution will occur during the bulk of days with low or moderate concentrations, and only a small additional contribution is to be expected from the few days with relatively high concentrations. In part, this also holds true if the low end of the exposure-effect relationship shows the non-linear shape described before.

2.2.3 Effects of mixtures of air pollutants

In chamber studies, single pollutant experiments are common, practicable and useful. In reality, however, the population is exposed to a mixture of different and numerous pollutants at concentrations varying with time and space. Associations found between health effects and air pollution are always associations with the whole mixture of gases and aerosols during the study period (and, in addition, may be influenced by the exposure before the study period). To a certain extent adequate statistical methods can be used to separate the influence of different factors contributing to the overall effect, but due to co-linearity on the one hand and interactions of the pollutants on the other hand even the best methods available have their limitations.

This is a well-known problem with respect to sulphur dioxide and particulate matter. However, many studies from the US suggest that relationships between health effects and pollution are much clearer for PM than for SO₂. On the other hand, the HEI study [37] and the results from APHEA [36] - as far as available at present - reinforce the importance of SO₂ as an indicator of the severity of an air pollution situation. Thus, the risk to be assigned to PM under "European conditions" might be lower than suggested by those studies (mainly from the US) in which PM was the only pollutant showing significant correlation to health effects. Multi-pollutant models should be able to provide more insight into mutual influences of PM and sulphur dioxide, but differences in the reliability of exposure estimates and nonlinearities can affect judgements about the relative importance of competing pollutants [71].

This situation is complicated by the fact that PM is not a single substance but as such also some kind of mixture of pollutants, in this case with differences in chemical properties and varying physical properties like size and surface, which greatly influence distribution and deposition in the respiratory tract. Some constituents of PM are closely linked with other pollutants, e.g., sulphur dioxide with sulphates, and there are suggestions that sulphur dioxide could be a surrogate for fine (acidic) particles. This further increases the difficulties of attributing health effects to single pollutants, and this also decreases the predictability of health benefits associated with abatement measures.

2.2.4 Relevant health endpoints and exposure-effects relationships

On an individual level, mortality is the most severe adverse health outcome. At a population level, however, other health effects may - in overall public health terms - be as important or even more important than mortality.

The significance of premature mortality depends on the degree of prematurity. If premature death refers to a few days of shortening of lives of individuals who are in such a state of ill-health that they are going to die with or without air pollution within the next few days, the public health importance is quite different from that of a situation with increased mortality in healthy individuals otherwise not expected to die during the next months or years. In the first case, a short-term increase in mortality is followed by a corresponding decrease; due to the depletion of the susceptible population (cf. section 2.2.5), this would not substantially change life expectancy in a population. In the second case, life expectancy might be markedly reduced, and this situation is clearly of much greater public health importance.

The data base concerning "short-term" exposure-effect relationships is much better with respect to mortality than other health endpoints because of the wide availability of mortality data and a lower probability of misclassification at least if total mortality is considered. This favours the use of mortality data in health effects assessment.

Other relevant health endpoints, the importance of which is not necessarily inferior to that of premature mortality, are hospital admissions, respiratory diseases and symptoms, or the need of additional medication in asthmatics. For these endpoints, health effects linked with air pollution are supposed to be

additional ones, at least to a considerable degree (i.e., in addition to the effects occurring in the absence of pollution and not only effects based on prematurity; therefore this increase is expected to be avoidable by abatement measures).

Thus, their overall public health impact may exceed even that of premature mortality. Finally, changes in lung function do not seem to be very sensitive health endpoints, and small transient changes are difficult to understand in their relevance to public health.

2.2.5 Exposure-effects relationships

It is obvious that the most susceptible individuals are those who will suffer first from mortality and morbidity, and therefore an increase in daily PM concentration is likely to be associated with an increase in daily mortality and morbidity. But what happens the day after an increase of PM concentration? If the concentration remains at a high level, yesterday's most susceptible individuals will already have experienced effects. In case no "new" individuals have entered the group of the most susceptible ones, the number of individuals at risk has decreased, and therefore the effects found will decrease, too (this concerns mortality, morbidity, hospital admissions, and symptom exacerbation, but not "repeatable" events like medication use and asthma attacks). If, on the other hand, new members have completely filled up the group of the most susceptible individuals overnight, the effect of the actual concentration of air pollutants will be the same as it was the day before, and in this case the effect will be directly related to the absolute concentration and not to the change. If the group of very susceptible persons has not been filled up, the acute effect will be much smaller and be related rather to the change and only in part to the absolute concentration. Probably the "real" situation is between these two extremes, i.e., the group of very susceptible individuals is filled up again with some lag in time. The interrelation resulting is outlined in Figure 2.2.

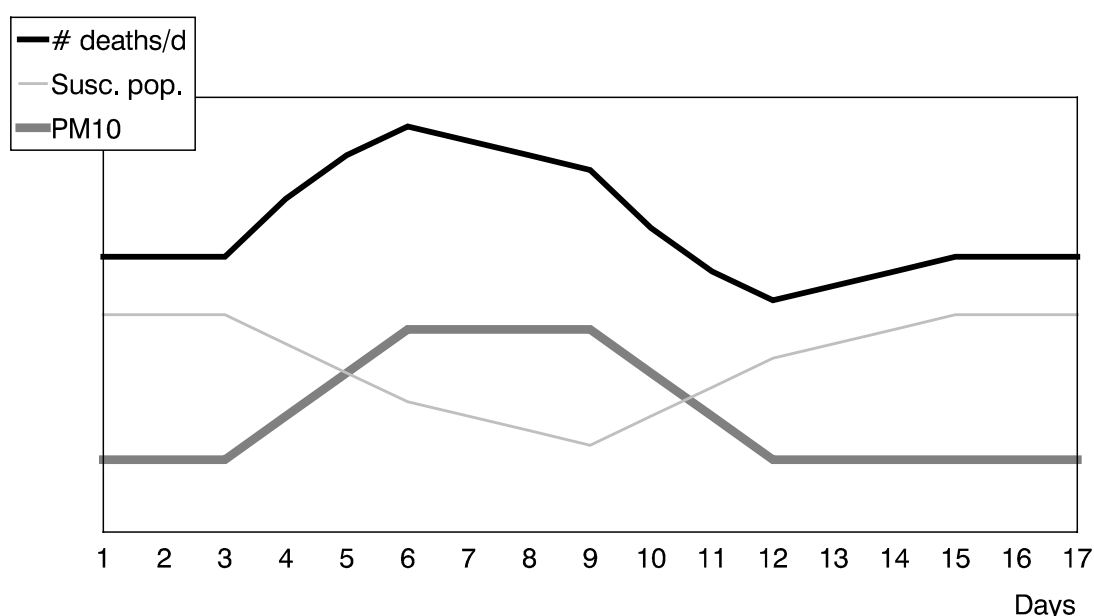


Figure 2.2: Hypothetical relations between changes in PM₁₀ concentration, size of susceptible population, and number of daily deaths.

In this schematic graph, constant PM₁₀ concentrations are supposed before and after the days considered.

A data set used in time-series analyses consists of all observations during the study period, i.e., pairs of exposure and effect data of all study days. Thus, a certain daily mean concentration will be linked with the health outcome found during increasing, constant, or decreasing concentrations, and in the end the exposure-effect relationship found is based on some kind of average of the slopes found at increasing, constant, and decreasing concentrations.

Relative Risk

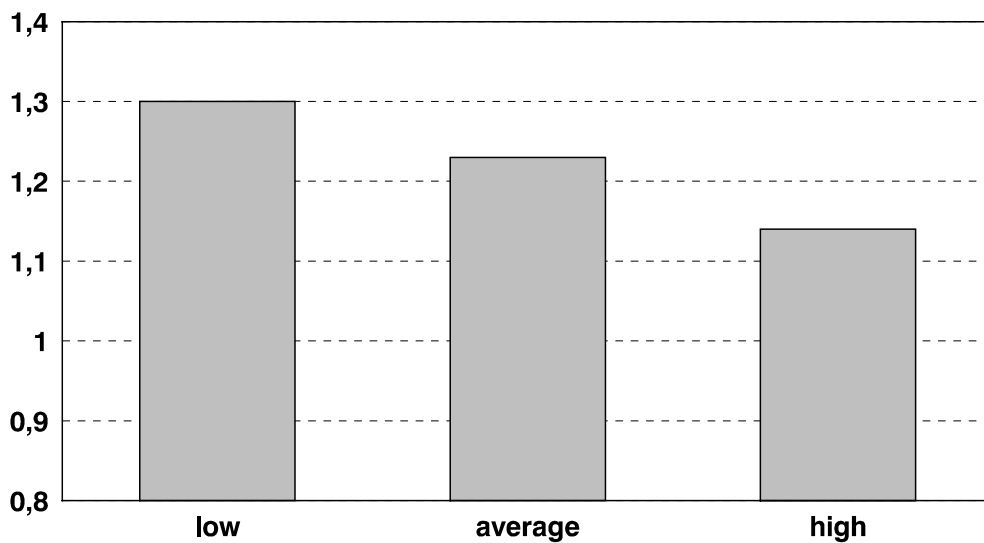


Figure 2.3: Changes of Relative Risk (RR) of daily mortality at different levels of mortality (low, average, high) during the previous 15 days.

RR is given in comparison of the 95th percentile vs the 5th percentile, i.e. 331 vs 15 $\mu\text{g m}^{-3}$ of SP. "Low" means that mortality during the previous 15 days was low, and the RR amounts to about 1.3 for an increase of 24h mean concentration from 15 to 331 $\mu\text{g m}^{-3}$. Adapted from [44].

This implies that in a period of increasing concentrations effects tend to be underestimated using the average slope; during a period of decreasing concentrations they tend to be overestimated. Findings from Erfurt (former GDR) support these considerations. The Relative Risk (RR) of daily mortality associated with a certain PM concentration was high after a period of low daily mortality (and low concentrations) and low after high daily mortality (and high concentrations) (cf. Figure 2.2).

There is another relevant implication. The slope S of the exposure-effect relationship shown in Figure 2.1 is:

$$(1) \quad S = (C-D)/(K-I).$$

Then the Total Health Outcome (THO) at day i with concentration c_i will be

$$(2) \quad \text{THO}_i = D + (c_i - I) * S,$$

and the total health outcome (THO) during a study period of n days will be

$$(3) \quad THO = \sum THO_i = \sum (D + (c_i - I) * S) = n * D + \sum ((c_i - I) * S) = n * D + S * \sum (c_i - I) \text{ for } i=1 \text{ to } n.$$

The mean concentration m during the study period of n days is

$$(4) \quad m = (\sum (c_i)) / n.$$

This leads to

$$(5) \quad \sum (c_i - I) = n * (m - I).$$

Thus (5) in (3) results in

$$(6) \quad THO = n * D + S * n * (m - I).$$

This means that the slope of an exposure-effect relationship derived from time-series analyses based on 24h data concerning concentrations and health outcome describes the slope of the relationship between **average** concentrations and effects rather than that between **short-term** changes and effects.

2.2.6 Results from APHEA and PEACE

PM is not a chemically defined homogenous substance but differs from site to site with respect to particle size distribution, components, and pattern of exposure. Therefore, epidemiological studies performed in the US may lead to results different from those based on European studies, and European results may be more appropriate in a European standard setting process; even though conditions may be different in different European countries with respect to exposure and other factors.

From the preliminary APHEA results, Table 2.3 based on time-series analyses can be derived.

Table 2.3. Preliminary APHEA results [39]

Health effect indicator	$\Delta C(\mu\text{gm}^{-3})$	$\Delta HI (\%)$	$\Delta C (\mu\text{gm}^{-3})$	$\Delta HI (\%)$
Daily mortality *	125	5	10	0.4
Hospital admissions for respiratory conditions **	100	5	10	0.5

* based on $RR=1.04$ for a $100 \mu\text{gm}^{-3}$ increase in PM_{10} (24h).

** based on $RR=1.019$ (15-64 years) and $RR=1.033$ (65+ years) for a $100 \mu\text{gm}^{-3}$ increase in TSP (24h); the total number of admissions was nearly the same for the 2 age groups, thus combined $RR=1.026$.

It has to be mentioned, however, that real PM_{10} measurements have not been carried out in APHEA. For some cities, PM_{13} and PM_7 data were available which have been used as a proxy for PM_{10} . Other calculations are based on TSP data converted to PM_{10} . On the other hand, a considerable part of the US time-series studies which refer to PM_{10} are based on conversions from PM metrics other than PM_{10} or not on daily measurements [69].

The time-series studies referred to in the WHO Working Paper (cf. Tables 2.1 and 2.2) show that the APHEA results suggest effects on daily mortality and hospital admissions for respiratory conditions which are in good accordance with the lower bound of the range [33, 69, 70]. A similar situation can be

stated for preliminary PEACE results with respect to bronchodilator use and symptom exacerbation among children with chronic respiratory symptoms.

2.2.7 *Effect estimates based on the results of time-series analyses vs. cohort studies*

Time-series analyses based primarily on short-term changes give no information on long-term effects. Two recent cohort studies, however, provide some information on combined short-term plus long-term effects of PM exposure on mortality [41, 42]. According to these studies, a $10 \mu\text{gm}^{-3}$ increase in annual mean PM_{10} concentration is associated with a relative risk of about 1.05. (The exact RR's for a $10 \mu\text{gm}^{-3}$ increase in annual mean concentration were 1.095 for PM_{15-10} based on a cohort size of 8,111 persons [41] and 1.069 for $\text{PM}_{2.5}$ based on a cohort size of 552,138 persons [42]. Considering an average $\text{PM}_{2.5}/\text{PM}_{10}$ ratio of 0.57 for the US [35] and giving more weight to the larger cohort, a RR of 1.05 per $10 \mu\text{gm}^{-3}$ difference in PM_{10} concentration seems to be a reasonable estimate.)

At first glance, this relative risk of 1.05 seems to be in contradiction to the findings from time-series analyses (i.e., it seems to suggest long-term effects in addition to short-term effects), which indicate an increase of daily mortality by 0.6 % per $10 \mu\text{gm}^{-3}$, i.e., a RR of only 1.006. The two sets of results cannot strictly speaking be compared. Since causal mechanisms are as yet not understood they may or may not address the same underlying phenomena. But the magnitude of the two sets of results is in fact comparable. In the time-series analyses the RR refers to daily mortality, i.e. the number of deaths per day in a population, whereas in the cohort studies the RR refers to the risk of an individual dying at a certain age. A daily RR of 1.006 per $10 \mu\text{gm}^{-3}$ means that the number of deaths per day is increased by 0.6% when concentrations of PM_{10} are elevated by $10 \mu\text{gm}^{-3}$ for one day. If concentrations were elevated above the normal by $10 \mu\text{gm}^{-3}$ every day for a year the same would hold true for the number of deaths per year. This may be illustrated by the following example. In a population of 100 000 individuals having a life expectancy of 80 years, $100\,000/80 = 1250$ will die per year. If this yearly rate is increased by 0.6 % to 1257.5, life expectancy amounts to $100\,000/1257.5 = 79.52$ years, i.e., a reduction of 0.48 years. If the relationship found in the two cohort studies is applied to life tables for Dutch or German men, a $10 \mu\text{gm}^{-3}$ increase in annual mean PM_{10} concentration corresponds to a reduction of life expectancy of about 0.5 years. This is in good agreement with the estimate via the time-series analyses and therefore does not necessitate (but also does not exclude) additional or different long-term effects on mortality. When the final version of the WHO report is available, further comments will be given on the effects of particles on life expectancy.

Figure 2.4 explains why age-dependency of mortality is an important reason for the numerical difference between RR derived from time-series analyses and RR derived from cohort studies. From a cohort of 100,000 persons followed from birth to death, the majority dies at higher age. Increasing the number of persons who die from birth to high age by 10% (RR = 1.1; at lower RR the two curves overlap widely) results in the small change of the curve of mortality shown in the upper part of Figure 2.4. (taking the example of German men). The age at which 50,000 of the 100,000 persons in the cohort have died shifts from 75.6 to 74.4 years, i.e. a difference of 1.2 years or 1.6% as a result of a RR of 1.1. However, if mortality were independent of age, i.e., the number of persons who die in a birth-to-death cohort would be the same every year, mortality would follow the linear course shown in the lower part of Figure 2.4. In this hypothetical case of age-independent mortality, the age at which half of the cohort have died would shift from 50 to 45.1 years, i.e. a difference of 4.9 years or 10% as a result of a RR of 1.1.

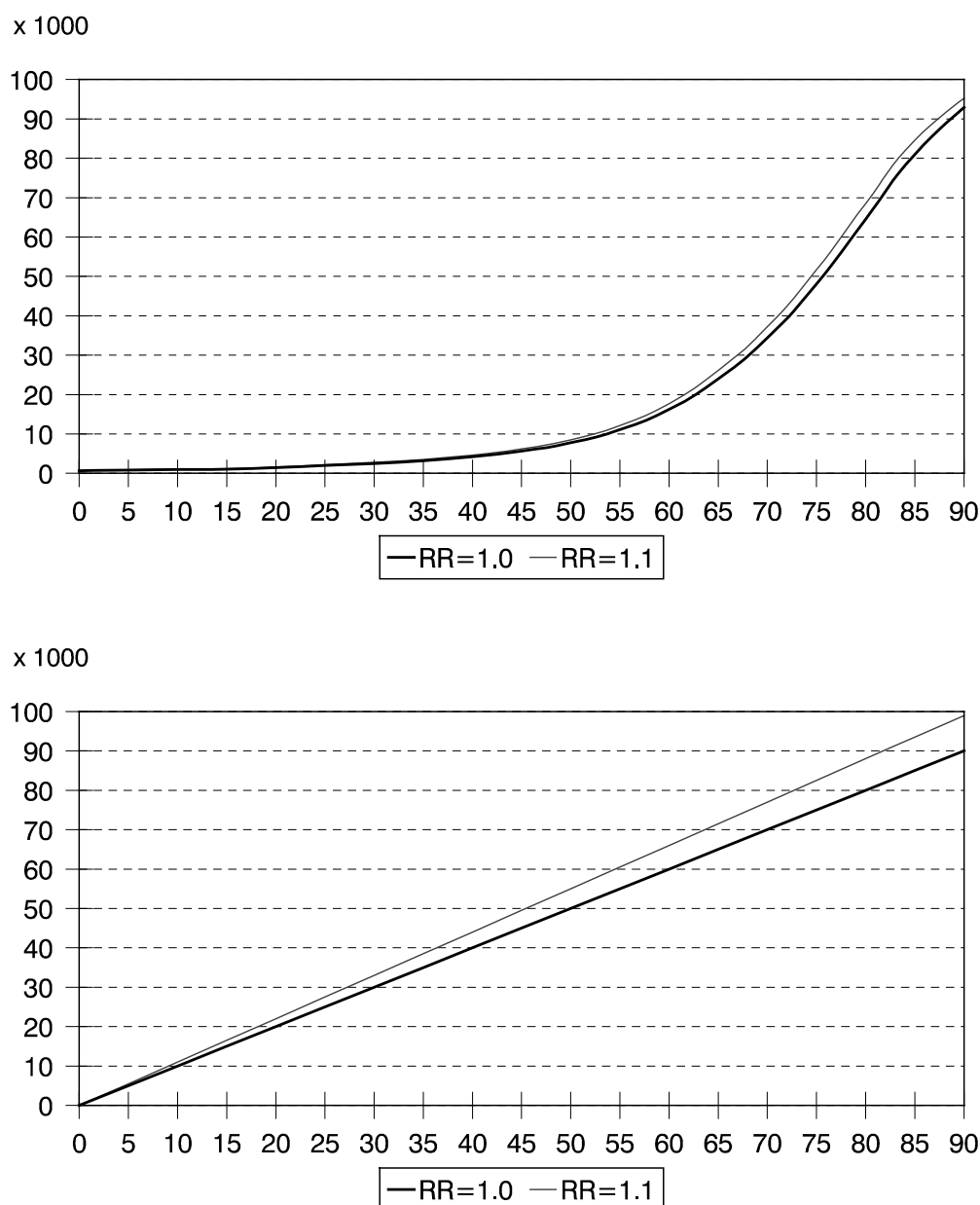


Figure 2.4: Influence of age-dependency on mortality in a birth-to-death cohort with a Relative Risk (RR) of 1.1. Upper part: "Real" mortality curve of German men [67]. Lower part: Hypothetical age-independent mortality curve.

Y-axis: Number of persons in a birth-to-death cohort who have died at the age (in years) indicated on the x-axis.

2.3 NUMBER OF INDIVIDUALS CONCERNED IN EUROPE AT THE PRESENT TIME

If there is no real threshold below which no health effects occur, the total EU population, i.e. about 370 million people, is potentially affected.

According to "Concern for Europe's Tomorrow" [66], 75 % of the population of western countries of the WHO European Region live in urban areas. 50 % of urban population with data available (24.6 million people in Western European countries) are exposed currently to annual TSP concentrations of $60 \mu\text{g m}^{-3}$ or more, which might correspond to some $40\text{--}50 \mu\text{g m}^{-3}$ PM_{10} .

If the sample of urban areas with PM measurements available is representative for all EU urban areas, at least 140 million EU citizens (50 % of 75 % of 370 million) are exposed currently to *mean* concentrations probably associated with health effects even when exceeded for 24 hours (cf. Section 2.4). Even if these concentrations are representative only for the 75 million people living in conurbations with more than 500,000 inhabitants, more than 30 million individuals would be exposed to these PM concentrations. Considering the relatively small differences between urban and suburban or rural PM concentrations (cf. PEACE results), a substantial percentage of EU citizens living in rural areas are exposed currently to PM concentrations probably associated with health effects, too. It should be noted, however, that PM concentrations are expected to improve (see section 1.8) and, therefore, the number of individuals affected is also expected to decline.

2.4 DEGREE OF EXPOSURE AND IMPACT OF POLLUTANT

Even if there is no "real" threshold, it is possible to gain some idea of the lower range in which the slope of the exposure-effect relationship decreases. In the HEI re-analysis of mortality data [37], study days of the different studies were classified by PM concentration quintiles (in Birmingham quartiles), and for each fifth (quarter) in this classification the RR of daily mortality was calculated separately. The RR in the class with the lowest exposure days was taken as a reference (RR=1.0). In most cases, the RR did not increase steadily with exposure concentrations, but was only increased for the highest concentration range. Table 2.4 shows the concentration separating the "normal risk" concentrations from the "higher risk" concentrations, i.e. the concentration at which the slope of the relationship between the concentration and the RR of daily mortality starts to increase ("change of slope concentration"). Below this concentration a linear concentration-effect relation between PM and mortality is very unlikely. Above this concentration, linearity is not demonstrated but may be a reasonable assumption. According to Table 2.4 the concentration in question might be near $40 \mu\text{g m}^{-3}$. At present, no comparable data are available from APHEA or PEACE.

Table 2.4: Change of slope concentrations for relationship between Relative Risk of daily mortality and PM concentration

Location	Philadelphia	St. Louis	East. Tenn.	Utah Valley	Birmingham
Study Period	73-80	85-86	85-86	85-86	85-88
HEI Tables no.	9/10	21/22	24/25	27/28	30/31
PM Measure	TSP	PM ₁₀	PM ₁₀	PM ₁₀	PM ₁₀
Median ($\mu\text{g m}^{-3}$) *	73	24	30	38	44
Change of slope concentration ($\mu\text{g m}^{-3}$)	95	40	(19) **	(42) **	45

* of all 24h values during study period

** increase in RR not significant

A similar concept is the "minimum clear increased risk" level which is used in a USEPA Office of Air Quality Planning and Standards (OAQPS) Staff Paper draft [45] Reconverted to PM₁₀ levels (24-hour average), these levels amount to 33 - 60 $\mu\text{g m}^{-3}$ for mortality, 19 - 52 $\mu\text{g m}^{-3}$ for hospital admissions, and 30 - 55 $\mu\text{g m}^{-3}$ for respiratory symptoms. These levels are in good agreement with the "change of slope concentration" in Table 2.4. (The OAQPS Staff Paper draft of April 1996 apparently has modified this argument and now uses the term "cutpoint" with a slightly different meaning [38]).

It should be emphasised, however, that these levels or concentrations do not demonstrate a threshold for acute health effects.

The following examples may give an impression of the health relevance of increased PM concentrations. They are based on the health effects estimates given in Table 2.1 (with the modification that a RR of 1.006 is used instead of 1.0071; the reason is that among the studies the WHO Working Paper is based on, there are two studies with averaging times longer than 24 hours and one study which refers to the elderly; excluding these three studies results in a RR of 1.006). Table 2.5 presents estimates of health outcome attributable to a 3-day episode with an increase of PM₁₀ concentration by 100 µgm⁻³, e.g., from 50 µgm⁻³ to 150 µgm⁻³. It should be noted that this is not an estimate of the effect of an episode in comparison to a theoretical zero exposure episode. These calculations are based on linear relationships, i.e., a 6-day period would result in twice these effects, an episode with a 50 µgm⁻³ increase in half of the effect of that with a 100 µgm⁻³ increase. This also means that the health effect attributable to an episode of increased air pollution equals the effect of the corresponding change in annual mean concentration induced by this episode, i.e., the health effect of a 100 µgm⁻³ increase lasting 3 days corresponds to the health effect of a 0.8 µgm⁻³ increase in annual mean concentration.

Table 2.5: Estimated number of cases attributable to a period of three days characterised by an increase in average PM₁₀ concentration from 50 to 150 µgm⁻³, adapted from [33]

Health endpoint	Number of cases (95% C.I.)
Mortality	6 (5-7)
Hospital admissions for respiratory conditions	6 (4-9)
Person-days of bronchodilator use	10110 (6150-14100)

Estimated number of cases in an example population of 1 million people, having on average 100 deaths and 75 respiratory hospital admissions per 3 days and on daily average 10,000 asthmatics using bronchodilators and/or experiencing asthma symptoms.

Based on the health effects estimates given in Table 2.2 (short-term exposure-response relationships) with the modification described for Table 2.5 and on equation (6) in section 2.2.5, Table 2.6 presents estimates of the increase in health outcome attributable to an increase in annual PM₁₀ mean. These calculations are based on linear relationships, i.e., other increases would be associated with proportional effects. It should be noted that this Table does not provide an estimate of the absolute effect associated with a given annual mean concentration in comparison to a theoretical zero exposure.

Table 2.6: Estimated increase in number of cases per year attributable to an increase in annual mean PM₁₀ concentration.

PM ₁₀ Annual Mean Increase	Mortality	Hospital admissions for respiratory conditions
From 15 to 20 µgm ⁻³	36 (29 - 43)	38 (23 - 53)
From 15 to 25 µgm ⁻³	72 (58 - 87)	77 (46 - 107)
From 15 to 30 µgm ⁻³	108 (87 - 130)	115 (68 - 160)
From 15 to 35 µgm ⁻³	145 (116 - 173)	153 (91 - 214)
From 15 to 40 µgm ⁻³	181 (145 - 217)	192 (114 - 267)

Estimated number of cases in an example population of 1 million people, having on average 33 deaths and 25 respiratory hospital admissions per day.

Based on the health effects estimates given in Table 2.2 (short-term exposure-relationships), Table 2.7 presents estimates of the increase of health outcome attributable to an increase in annual PM_{10} mean. It should be noted that this Table does not provide an estimate of the absolute effect associated with a given annual mean concentration in comparison to a theoretical zero exposure.

Table 2.7: Estimated increase in number of person-days per year attributable to an increase in annual mean PM_{10} concentration.

PM_{10} Annual Mean Increase	Bronchodilator use among asthmatics
From 15 to 20 $\mu g m^{-3}$	61,500 (37,400 - 85,800)
From 15 to 25 $\mu g m^{-3}$	123,000 (74,800 - 171,600)
From 15 to 30 $\mu g m^{-3}$	184,500 (112,200 - 257,400)
From 15 to 35 $\mu g m^{-3}$	246,000 (149,600 - 343,200)
From 15 to 40 $\mu g m^{-3}$	307,500 (187,000 - 429,000)

Estimated number of person-days in an example population of 1 million people, having on daily average 10 000 asthmatics using bronchodilators and/or experiencing asthma symptoms.

As chronic effects are not explicitly included, there may be considerably more health effects than described in the Tables 2.6 to 2.7.

2.5 PRESENCE OF SENSITIVE SUBGROUPS

Aged individuals suffering from cardiorespiratory diseases are a sensitive subgroup of increasing importance due to the changing age distribution in the European population with a shift to older ages.

Asthmatics of all age groups are sensitive to PM-related health effects, too. Asthmatics represent a few percent of the population, but the prevalence is increasing.

In general, all individuals additionally exposed to PM for other reasons (active and passive smokers, occupational exposure) are expected to be more at risk than others.

2.6 HIGHLIGHTING OF DIFFERENCES BETWEEN MEMBER STATES

Some important differences between Member States are due to geographical factors and climate. The relevance of natural sources contributing to PM exposure is likely to be different between Southern and Northern Europe. The Mediterranean countries are much closer to the Sahara desert and therefore are more concerned with problems of long-range transport of airborne material of this origin. In wet and cold regions, resuspension of settled dust is less important than in dry and warm regions. In addition, grassland or woodland reduce rather than increase PM concentrations. These natural factors contribute to the distribution found in Europe with low PM concentrations in the North and relatively high concentrations in the South.

Personal exposure is also influenced by the percentage of time spent outdoors and by the air exchange rate in indoor environments which are higher in warm regions and thus tend to increase exposure to outdoor levels of particulates. On the other hand, better ventilation may decrease exposure to pollutants arising from indoor sources, e.g. to environmental tobacco smoke and to all pollutants with indoor sources.

2.7 RECOMMENDATIONS FOR LIMIT VALUES AND ALERT THRESHOLDS

2.7.1 *An appropriate measure of particulate matter*

A crucial point is the question of causality in the associations found between particles and the adverse health effects. In a strict understanding, epidemiology is only able to demonstrate statistical associations and not causal ones, but there are criteria (Bradford Hill) for assessing the likely causality of a relationship (strength of association, consistency, specificity, relationship in time, biological gradient, coherence, and - but this is not a necessary condition - biological plausibility). However, even if there is reason to believe that the associations between air pollution and health effects are causal, the problem of finding the causal agent (or even the best indicator) itself remains unsolved. This holds true for gaseous pollutants and aerosols in general and also for fractions of PM with different particle size.

Associations have been found or suggested to exist between health effects and BS, TSP, PM₁₀, PM_{2.5} and sulphates. TSP includes a considerable fraction of particles too large to enter the deeper parts of the respiratory tract. BS measures a property of particulate matter (blackness) linked with mass concentration in an unclear way which may change significantly from site to site according to the local mixture of sources. Therefore TSP and BS no longer seem to be adequate measures for particulate matter on which to base PM limit values. While TSP and PM₁₀ have been measured directly in a number of studies, this is usually not the case for smaller PM fractions. Instead, indirect estimates were used based approximate relations between PM₁₀, PM_{2.5} and sulphates or between haze and PM_{2.5} or similar relations which are known to be site-dependent. Therefore, it is difficult to decide which measure is appropriate.

However, there are some reasons to suppose the smaller PM fractions to be more closely linked to many health effects, at least to mortality:

- If there are statistical associations between effects and exposure to, e.g., TSP, PM₁₀, and PM_{2.5} (this has been demonstrated in some cases), then it is more convincing to assume that the smallest fraction is the driving force and not the larger ones (because the smaller fractions are components comprised within the coarser ones, but not vice versa).
- In general, people spend most of their time indoors, but correlations found refer to outdoor concentrations. The smaller the particles, the better the indoor/outdoor exchange; this provides a good reason for assuming smaller particles being involved in causality. Very fine particles show a much more uniform local distribution than the coarser ones. Thus outdoor measurements are a better proxy for individual exposure to smaller than to larger size fractions of PM. This should increase the probability of correlations between health effects and outdoor concentrations.

A mechanism by which PM may induce health effects is alveolar inflammation. This has been shown by Oberdörster exposing rats to 20 nm particles which induced effects (including mortality at relatively low concentrations) much greater than expected from their mass concentration [40]. A recent hypothesis [43] tries to link effects on the respiratory tract and cardiovascular effects. Acidic particles of less than 100 nm in diameter are supposed to provoke alveolar inflammation which causes both acute changes in blood coagulability and release of mediators able to provoke attacks of acute respiratory illness in susceptible individuals. The blood changes result in an increase in the exposed population's susceptibility to acute episodes of cardiovascular disease; the most susceptible suffer the most.

Bearing in mind:

- that, with respect to measurement equipment, intercomparison campaigns are being performed for PM₁₀, but at present not for PM_{2.5} or other fine particles fractions,

- the fact that a very important part of health effects studies are based on TSP or PM₁₀ measurements,
- and that, last but not least, PM₁₀ concentration levels show an acceptable correlation to PM_{2.5} levels,

PM₁₀ seems to be a reasonable compromise between theoretical arguments favouring the measurement of very small particles and the use of knowledge and practical experience based on PM₁₀ and TSP (and BS) measurements. Thus, if only one PM fraction is to be selected, at present PM₁₀ would be an adequate choice. However, in a few years there might be the need to add PM_{2.5}, another fine particles fraction, or properties other than mass such as number or surface of particles, depending on new effects data and the availability of adequate and comparable measurement equipment. For this reason, it is recommended that PM_{2.5} measurements are carried out at selected sites in order to gain a greater understanding of its phenomenology. Advances in understanding of the mechanisms of the toxicological action of particles may also provide information on which size fraction, or whether the number, surface area, mass or composition of particles is important.

2.7.2 Averaging periods

Health effects studies are very often based on time-series analyses. In the case of PM, 24 hours are a common and useful averaging time period describing short-term changes in both exposure and effects. Present knowledge is mainly based on exposure-effect relationships with a temporal resolution of 24 hours or simple multiples of 24 hours. This does not necessarily mean that effects are "really" related to 24h mean concentrations (instead of shorter peaks occurring during the averaging time), but at present 24h means seem to provide an acceptable description of outdoor concentration patterns. In addition, annual means or similar data (e.g., median) derived from 24h means provide useful information on distribution of 24h values over a longer time and on average long-term exposure and are necessary to assess health effects.

There are also relationships between annual means and upper percentiles of daily means. At a given annual mean, the 98-percentile of 24h concentrations amounts to two to three times the annual mean concentration as suggested by Tables A 2.1 to A 2.10.

2.7.3 Alert Levels

Supposing a linear (or, at higher concentrations, a sub-linear) concentration-effect relationship without a threshold, no alert values can be proposed on a scientifically sound basis. However, alert levels may be meaningful under special local conditions; in this case, local decisions on alert thresholds and adequate measures to be taken in case of alert seem to be more useful than EU-wide regulations. In general, abatement measures known to effectively lower annual mean concentrations and peak concentrations should be sufficient to protect the population from unacceptable risks to health.

2.7.4 Limit Values

The following basic statements can be made:

- At present, limit values should be derived for PM₁₀.
- The relation to health effects is not so clear for TSP and BS, thus these metrics should not be used for defining limit values in the Directive. However, further research on health impacts of

BS and on the PM fraction related to BS measurements is justified. Also, scientific reasons suggest that these measurements should not be stopped completely at locations where they have been carried out in the past.

- For reasons of practical feasibility, the shortest averaging time of the limit value should be 24 hours.
- Good reasons might be given for considering other fractions rather than just PM_{10} , e.g., $PM_{2.5}$, and there might be an increasing need of such kind of measurements in the future. At present knowledge on health effects of particle fractions is insufficient, and sufficiently standardised measurement methods are not available to provide a sound basis for limit values for particle fractions smaller than PM_{10} . As this will probably change in the future, limit values set for PM_{10} now may have to undergo revision at a later stage.

Limit values should be set for an annual mean combined with an upper daily limit value.

For practical reasons, absolute upper limits are not very adequate. Therefore an upper limit (24h) should be derived with very few exceedances allowed, defined, e.g., as a low number of accepted exceedances per year (in the range of 2 - 7) directly indicated or as a 98 to 99.5 Percentile. The final choice of the percentile may be influenced by the better stability of the 98-Percentile. In any case, a set of limit values consisting of an annual mean and a maximum daily mean with a low number of annual exceedances allowed should be balanced, i.e. the relation between these two limit values should reflect a "normal European situation" in the sense that control measures are more or less equally necessary to meet both limit values. Nevertheless, under specific local conditions one of the two limit values may be more difficult to meet than the other one. Reference to a "normal" relation between the two limit values should help to avoid dramatic changes in the composition of the aerosol which might be induced by measures predominantly oriented either to lower the annual mean (and accepting high peaks) or to diminish peak concentrations without lowering the annual mean. However, measures inducing a significant change in aerosol composition and size distribution might lead to unpredictable results with respect to health effects.

The fundamental definition of limit values is ultimately a political decision which should be taken with regard to the estimated health effects given in Section 2.4. As chronic effects are not explicitly included, there may be considerably more health effects than described in this chapter.

Assuming that there are no thresholds below which health effects are not to be expected, no limit values can be proposed that are strictly based on scientific arguments. However, it seems reasonable to suppose health effects to occur at least at 24h concentrations as low as $40 \mu g m^{-3}$ and even down to background concentrations. Therefore the Working Group is of the opinion that PM_{10} limit values should be set within the following ranges: $30-100 \mu g m^{-3}$ as a 24h average concentration and $15-40 \mu g m^{-3}$ as an annual mean concentration.

There was intensive discussion in the Group on whether a single number could be given rather than a range. Arguments were brought forward against the Group setting a single number. These included: that the uncertainties were so great that to recommend a single value would suggest a false certainty; that the final outcome from the WHO AQG update was not yet available; and the lack of information about achievability of cost. The point was made, and accepted, that there must be a gap between the natural background level and any limit value recommended by the Group. The majority of the Group agreed that a 24-h average limit value could be around $50 \mu g m^{-3}$. The arguments for this number were as follows:

Using the information given in sections 2.1 and 2.4, a rise from a daily average PM_{10} level of 20 to $50 \mu g m^{-3}$ would be expected to be associated with between $\frac{1}{2}$ and 1 extra hospital admission daily for respiratory disorders in a population of 1 million. A recent study in Birmingham, UK (population 1

million), which the UK Expert Panel on Air Quality Standards considered in recommending a standard of $50 \mu\text{g m}^{-3}$, showed a similar result with an increase of just over 1 hospital admission daily for respiratory disorders for the same increase in PM_{10} levels [78]. Moreover, the number is also of similar magnitude to the “change of slope” arguments discussed in Section 2.4. Finally, the value of $50 \mu\text{g m}^{-3}$ as a 24-hour average has been agreed as a target for particulate matter in the Auto-Oil study. While using the latter argument as a rationale for the recommendation is to some extent circular, there is nonetheless a considerable amount of useful information on the achievability of a $50 \mu\text{g m}^{-3}$ 24-hour average value in the Auto-Oil study.

Some qualifying comments were made to put this number in perspective. In particular the question of geogenic dust which plays an important role in some Member States, has not been taken into account and needs further consideration by the Commission.

Concern was also expressed that money needed for technical measures to perhaps only achieve a small reduction in PM_{10} levels may be better invested to lower other (more important) health risks. However, it was also emphasised that control strategies other than technical ones are applicable.

Taking all these considerations into account, on balance the majority of the Group recommended a 24-hour PM_{10} limit value of $50 \mu\text{g m}^{-3}$ as a 98-Percentile (of daily mean values over a calendar year) in conjunction with an annual mean limit value of $20 \mu\text{g m}^{-3}$, on the basis that typical ratios between 98-Percentiles of daily means of PM_{10} and annual averages are about 2.5. The Group came to this view primarily on the basis of a consideration of health effects, although the results of the Auto-Oil study gave some indications of achievability of this value. The Group puts forward this recommendation recognising that the project on economic evaluation has still to report fully, and that the Group's recommendation will be assessed in the light of the economic evaluation, by the Steering Committee. On the basis of the epidemiological studies referred to earlier, it would be expected that adverse effects could still occur below the recommended limit values.

Setting relatively low values that are very difficult to meet at present and therefore requiring a large margin of tolerance may be useful in combination with a time-table defining steps concerning size and time of reducing this margin of tolerance.

Whatever decisions are finally taken on the values, the Group recommends that they be reviewed within 5 years after entry into force. It is assumed that after that time more scientific information will be available, also with regard to the question as to whether PM_{10} , $\text{PM}_{2.5}$ or another parameter is the appropriate indicator for exposure to suspended particulate matter.

2.8 RESEARCH NEEDS

Exposure-effects relationships based on European data seem to differ to some extent from those indicated by US data; the slopes of these relationships tend to be lower in Europe. This might be due to a difference in the mixture of air pollutants or in the composition of particles in Europe and in the US. First results from APHEA and PEACE have confirmed the necessity of European limit values to be based on European data.

To further clarify this situation, research directed to air pollution epidemiology, exposure assessment, and air pollution health effects assessment should be encouraged and supported in Europe in order to improve the database for setting European standards appropriate to the European air pollution situation. This is necessary for future decisions on adequate PM measures and future revisions of air quality standards with respect to PM-related health effects.

There is also the need for further research with respect to the causality of short-term and long-term exposure-effect relationships. This is closely linked with the question as to what are suitable PM/PM₁₀ abatement measures and whether these measures will result in an adequate reduction of health risk.

2.9 SUMMARY OF KEY POINT AND RECOMMENDATIONS

There is increasing evidence that health effects occur at very low levels of Particulate Matter (PM) and without an apparent threshold. Exposure-effect relationships found in recent European epidemiological studies are in good accordance with the lower end of the range found in US studies.

For the time being, PM₁₀ as a PM metric seems to be a reasonable compromise between theoretical arguments favouring the measurement of small particles and the extent of knowledge and practical experience based on PM₁₀ and TSP (and BS) measurements.

24h-means provide an acceptable characterisation of outdoor PM concentration patterns. 24h limit values should be complemented by annual mean values.

The assumption of the absence of a threshold implies that, due to their number, days with low or moderate concentrations have the highest impact on the occurrence of adverse health effects; the contribution from the small number of high pollution days is relatively small.

As an effects threshold cannot be defined, the setting of limit values is ultimately a political decision which should be based on best estimates of exposure-response relationships under European conditions. However, the majority of the Group felt that it was possible to recommend levels at which effects on public health would be small.

Without an effects threshold, no alert values can be proposed on a scientifically sound basis. If, nonetheless, for specific reasons alert values are considered useful for some areas, local decisions on alert thresholds and adequate measures to be taken in case of an alert seem to be more useful than EU-wide regulations.

The Working Group recommends that PM₁₀ limit values should be set within the following ranges: 30-100 µgm⁻³ as a 24h average concentration and 15-40 µgm⁻³ as an annual mean concentration. Within these ranges, the majority of the Group recommend a 24-hour PM₁₀ limit value of 50 µgm⁻³ as a 98-Percentile in conjunction with an annual mean limit value of 20 µgm⁻³. The Group puts forward this recommendation recognising that the project on economic evaluation has still to report, and that the Group's recommendation will be assessed in the light of the economic evaluation, by the Steering Committee.

Whatever decisions the Council takes finally on the values, the Group recommends that they be reviewed within 5 years after entry into force.

3. MEASUREMENT

3.1 INTRODUCTION

There is increasing recognition of the need for concerted and effective action to improve air quality. Air pollution is becoming of major influence on the quality of life, posing a risk both to human health and to the environment. In order to develop appropriate air quality management plans, however, it is necessary first to have reliable information about the state of pollution, a point recognized in the AGENDA 21 of the United Nations Conference on Environment and Development (UNCED) held in Rio de Janeiro in June 1992 (Annex 3).

The EU Directive on ambient air quality assessment and management states that when limit values (and alert thresholds) are set, ambient air quality shall be assessed in the Member States. Assessment is required throughout the territory of each Member State, but monitoring is mandatory only in areas with a high population¹, or pollutant concentration. If the concentrations are low, a combination of measurements and models may be used. However, for a complete assessment of the air quality situation, monitoring on a combination of spatial and temporal scales as well as modelling is necessary [46].

This Chapter sets out methods for air quality assessment and monitoring, and proposes a methodology to determine areas where monitoring is required and the number of stations to be established.

The compilation of an assessment of air pollution is difficult. The available data sets on the pollutants and their health and environmental impacts are often incomplete and sometimes out of date [47]. There are difficulties in compiling the data for a report on a European scale because of differences in methodology and reporting between countries and even within countries and cities. Shortcomings in the data which were used, including problems of representativeness and comparability of different data sets need to be addressed.

Monitoring stations provide data about air pollution levels at the specific time and specific site of sampling. Yet, as air quality is a highly complex spatial and temporal concentration field a sparse set of observations at several place-time co-ordinates cannot necessarily give an accurate picture of the extent and severity of the (urban) air pollution problem in an area. Even highly developed countries are limited in the number of observation sites that can be maintained due to the high cost of the sophisticated equipment required.

Recent experiences in several cities and countries showed that monitoring data can be complemented with spatial interpolation techniques, model calculations (dispersion models at different scales, deposition models, effect models etc.) and emission estimations and projections. Models allow the extension of monitoring data and to dynamically estimate ambient air pollution levels and depositions over differentiated emission and receptor areas. Some of these models, which may be combined with Geographical Information Systems (GIS), can be very simple and aggregated; others may be very sophisticated. However, even simple models complement monitoring data and are a valuable tool to assess effects of air pollution and develop the most effective abatement strategies.

For an assessment to be effective it must provide for the interpretation of air quality data in terms of the health of the residents of the monitored areas. Assessment of air quality for public health purposes, at the

¹ Article 2 states: A zone with a population concentration in excess of 250,000 or, where the population concentration is 250,000 or less, a population density per km² which for the Member States justifies the need for air quality to be assessed and managed.

moment, consists essentially of examining the prevalent air quality against established guidelines in terms of extent and number of exceedances, area of exceedance and number of people exposed.

3.2 MEASUREMENTS OF FINE PARTICULATES

3.2.1 *Introduction*

This Section discusses and describes monitoring techniques appropriate for fulfilling the requirements of the Directive. Generic measurement techniques for particulate matter, appropriate to the time scale of the proposed limit value, are discussed, and a system of traceability to reference monitoring methods and QA/QC procedures required to ensure high quality data are described. Proposals for determining areas where monitoring is required and the number of monitoring stations to be established are presented.

3.2.2 *Measurement methods*

This section gives a general overview of methods of measurement of particulate material in the atmosphere. It is not intended to be exhaustive or to give full operational details.

Sampling instruments for the gravimetric determination of airborne particles consist of a sample inlet, collection medium and a flow regulated pump.

3.2.2.1 *Sample inlet*

The sample inlet defines the size range of the particles collected. In broad terms, the normal size ranges sampled are TSP, PM₁₀ or other fractions of TSP. In practice, most TSP inlets in fact have a particle size cut-off limit, which may or may not be well defined. PM₁₀ sample inlets are generally based on a limited number of commercial designs, whose characteristics have been well defined in wind tunnel and ambient field tests. TSP sample inlets generally operate with high ($\approx 100\text{m}^3/\text{hr}$), medium ($\approx 10\text{m}^3/\text{hr}$) or low flow rates ($\approx 0.083\text{m}^3/\text{hr}$). Commercial PM₁₀ sample inlets are available for high, medium and low volume samplers. The performance of many of these sample inlets has been tested in US and EU comparison programmes and reports of these studies should be consulted for further details [29, 48 - 52].

Recently, specific PM_{2.5} sampling heads have become available for commercial low volume samplers. However, at present, fine particulate material like PM₅, PM₁ or PM_{0.5} is generally determined by cascade impactors with high or low flow rates.

Monitoring of particulate material by the black smoke method (ISO 9835) [8, 9] is also widely undertaken in many Member States. Although there is high correlation between black smoke and PM₁₀, the ratio between these two parameters varies from location to location and season to season, depending on the source of the particles monitored. Therefore, black smoke being based on the principle of reflectance measurement, is no measure for PM₁₀ or other gravimetrically determined particulate matter.

In optical monitoring devices, a TSP sample inlet may be used, with size fractionation undertaken at the (optical) analysis stage. Nephelometers are primarily only useful for measuring very fine particles (PM_{<2.5}).

To ensure correct operation, all sample inlets and sample lines, especially size selecting inlets, require regular maintenance (for example cleaning of sample lines and greasing of some inlets) in accord with the manufacturers instructions.

3.2.2.2 *Collection medium*

In most instruments, the particulate material is collected on a filter for either on-line or off-line weighing. The choice of filter material type is not generally critical, unless the filters are to be used for subsequent chemical analysis in addition to weighing. (This may be relevant when consideration is given to measurement of the compounds Pb, Ni, Cd and As, within the Framework Directive). However, for accurate mass determination of filters by manual off-line methods, conditioning of the filter before pre- and post- weighing is essential to ensure that moisture absorbed by the filter does not act as an interferent in the determination of the mass of material collected.

In general, quartz or glass fibre filters are recommended, rather than cellulose, and the filter conditioning procedures given in the current Council Directive 80/779/EEC on air quality limit values and guide values for sulphur dioxide and suspended particulates (24-hours at 20 °C and 50% relative humidity).

Conditioning of the filter in automatic on-line weighing analysers may also be an issue. Some analysers (e.g. TEOM) maintain the filter at 50 °C, thereby ensuring constant filter conditions. However, this is likely to remove water and some volatile hydrocarbon species and appears to lead to occasional recording of short term negative mass concentrations. Heating of the sample inlet for automatic analysers may be necessary for some automatic analysers, to avoid water condensation on the filter. Some on-line analysers do not maintain constant filter conditions (e.g., some β -absorption instruments) and hence, different relative humidity conditions may lead to some variability between measurements, particularly between summer and winter [53]. Therefore, it is recommended that these instruments are operated in a temperature controlled environment.

3.2.2.3 *Mass Measurement*

Measurement of the mass of material collected can be undertaken by direct weighing either off-line or on-line. Alternately, surrogate methods of mass determination, such as β absorption can be used.

In the black smoke method, measurement is made by determination of the reflectance of the filter stain by a calibrated reflectometer.

The following tables (Table 3.1 and Table 3.2) list the monitoring instruments currently in use in national networks in Member States. These data have been gathered from a questionnaire distributed by the Working Group. Table 3.1 shows that the β absorption and TEOM instruments are the most widely used for PM₁₀ monitoring. These techniques are also used for TSP monitoring but, in addition, some manual methods, such as the small filter unit and the HiVol sampler are also used. PM_{2.5} is monitored routinely in only one country, Sweden, but black smoke is still monitored widely throughout the EU.

Table 3.1: Measurement methods currently used in member states - PM₁₀ and TSP

MS	PM ₁₀		TSP	
	Method (averaging time)	Reference method	Method (averaging time)	Reference method
A	-		β-gauge (30min) ^a TEOM (30min)	
B	β-gauge (30min)		β-gauge (4h) nephelometry (30min) ^a	
D	-		β-gauge (24h) Small Filter and LIB Filter Device (24h random)	
DK	-		filter sampler (24h)	
E	β-gauge monitor (1h) TEOM (1h) gravimetry (24h)	US EPA	β-gauge monitor (1h) TEOM (1h) gravimetry (24h) nephelometry (1h)	US EPA
EL				
F	β-gauge (2h) TEOM (2h) optical (2h)		-	
I	-		method unknown	
IRL	-		-	
LX	TEOM (30min)		-	
NL	FAG-FH62IN (1h)		-	
P	β-gauge (4h)		High volume sampler (24h) ^a β-gauge (4h)	
S	TEOM (24h)	US EPA	-	
SF	Impactor (24h) Hivol sampler (24h) ^a TEOM (1h) β-gauge (1h)		Hivol sampler (24h) ^a Impactor (24h) Low volume filter sampler (1mth)	
UK	TEOM (1h)		-	

^a Most widespread measuring method.

Table 3.2: Measurement Methods currently used in Member States - PM_{2.5} and BS

MS	PM _{2.5}		Black Smoke	
	Apparatus (averaging time)	Reference method	Apparatus (averaging time)	Reference method
A		-	-	
B		-	Reflectometer (24h) β-gauge for PM ₁₀ (30min)	
D		-	-	
DK		-	SF8 (24h)	80/779/EEC
E	-	-	Low volume sampler (24h)	80/779/EEC
EL	NO DATA		NO DATA	
F		-	Filtromat (24h) Opale Cosma (24h) SF8 with total acidity (24h)	
I	NO DATA		NO DATA	
IRL		-	Reflectometer (24h)	BS1747 OECD curve
LX		-	SF8 (24h)	OECD
NL		-	OECD 1964 method (24h)	OECD
P	-		-	
S	Virtual impactor (48h)		OECD 1972 method (24h)	OECD
SF			-	
UK	-		Daily sampler (24h)	80/779/EEC

3.2.3 *Reference Sampler & Equivalence Reference Samplers*

Following the procedure adopted in previous EC Directives, it is proposed that a reference sampler be nominated. For monitoring compliance with the Directive limit value, Member States may then use the reference sampler or any other sampler which has been demonstrated as being equivalent to the reference sampler.

The Commission has undertaken an extensive series of studies on the development of a reference method for SPM monitoring [50]. Much of this work forms the basis of a series of papers and draft CEN standards produced by CEN/TC 264/Working Group 6 [51]. Based on the work of this group, the Wide Range Aerosol Classifier (WRAC) should be selected as the reference sampler for airborne particles. WG6 selected the Wide Range Aerosol Classifier (WRAC) as a reference sampler, for the following reasons:

- Presently, WRAC is the only system sampling both total and thoracic particles isokinetically at the same time from the same air flow in the central inlet system; it shows optimal sampling performance as compared to all other current sampling systems. This sampling process is based on established first physical principles, regarding the sampling efficiency of the central inlet as well as the aerosol impaction mechanisms.
- The WRAC system has been amply tested within the context of the program of the European Union to develop a reference method for suspended particulate matter [29, 49, 50, 52].
- The position taken by the EU, implying that the WRAC can be considered as provisory reference instrument for the measurement of thoracic particles within the framework of the forthcoming revision of the pertinent Air Quality Directive on SPM.

However, WG 6 recognise that there are severe limitations to the use of the WRAC, even as a reference sampler, due to its impracticability. The group therefore recommends that samplers that have been tested and shown to be equivalent to the WRAC, using the detailed field testing procedure documented by the group [51], can be designated as "equivalent reference samplers".

In view of the designation of the equivalent reference samplers as well as for the judging of sampling inlets, the CEN standard will set acceptability criteria for repeatability (from parallel measurements with two instruments of the same type) and for comparability (comparison with the reference sampler or one of the designated equivalent reference samplers).

Full details of the data evaluation procedure and acceptance criteria will be given in the standard.

The standard specifically relates to comparison of PM₁₀ inlets, rather than complete instruments and hence, suggested acceptance criteria presented in the standard do not include any allowance for differences in collection or mass determination of particulate material. Hence, when using the methodology of the standard to compare complete analysers (as used in monitoring networks) with the reference sampler or one of the equivalent reference samplers, a wider range of acceptability may be appropriate.

The Commission is currently undertaking a series of field intercomparisons in order to check the performances of complete instruments. These comparisons include instruments such as the β attenuation and TEOM analysers as well as new instruments based on other measurement principles.

With regard to fine particulate material, commercial PM_{2.5} inlets are available for low volume samplers, but these have not been fully tested and reference samplers have not yet been designated within the EU.

Black smoke monitoring cannot be used for determining compliance with the Directive but, where black smoke measurements are continued to maintain historical datasets or to assist in assessments, then monitoring should be undertaken by the methodology documented in the OECD report [8] and ISO 9835 [9], following the siting and operational procedures documented in the EC Instruction Manual for Black Smoke Monitoring [54].

3.2.4 Number & frequency of measurements

In general, to fully characterise a time series of particle concentrations, continuously operating monitoring devices are necessary.

However, for manual methods, the cost implications for daily sampling, for compliance checking may be unnecessarily high and more limited sampling may be acceptable. For example, in the USA, “24hr samples are collected at a frequency of every day, every other day or every sixth day depending on the conditions in the particular monitoring area” [55]. In Germany, sampling is often undertaken on a random basis using a grid pattern of sampling points throughout an urban area. Under this scheme, 120 24-hour samples per year are taken in areas where the burden is >80% of the limit value and 60 samples per year in other areas.

Data from Germany and the UK, including years with smog episodes such as 1987, show that random sampling at a single location on at least 60 days per year for mean values and at least 120 days per year for high percentiles gave good agreement with continuous methods within the range of the measurements.

Therefore, random sampling can be regarded as equivalent to continuous monitoring with the provision that:

- (i) the random sampling be evenly spread throughout the year
- (ii) the accuracy, with respect to continuous monitoring can be shown to be within $\pm 10\%$
- (iii) some degree of continuous monitoring is undertaken, for example at least one site per zone or agglomeration.

In the case of continuous automatic monitoring, the data capture should be at least 75%, with any missing data distributed throughout the year, rather than concentrated in an extended period. However, in the case of intermittent manual or random sampling, a data capture of at least 90% of the planned measurements should be achieved.

In areas where monitoring is not mandatory, lower data capture may be acceptable. However, in order to operate an alert warning system, if required, very high data capture from on-line automatic analysers is required. Data from such instruments are normally averaged into half-hour or hourly concentrations and such data will need to be available throughout the year. If required, 24hr rolling averages can be calculated from hourly data.

3.2.5 Network Design

Network design and site selection are fundamental in providing a harmonised and comparable system for measurement in each Member State, as well as being necessary and integral components of any quality assurance plan for particle monitoring.

In general, there are no hard and fast rules for network design, since any decisions made will be determined ultimately by the overall monitoring objectives. In practice, the number and distribution of air quality monitoring stations required in any extended network depends on the area to be covered, the spatial variability of the pollutants being measured and the required data usage. If identification or quantification of public health hazards associated with air pollution is an important factor, then the network design should consider the needs of, and information from, epidemiological studies. This may require site or pollutant specific approaches.

A general treatise on the design of air quality monitoring networks which satisfy the requirements of the EC Framework Directive has been provided by the Commission [46]. Clearly, the starting point for the development of the design of the PM₁₀ monitoring network is the legal requirements of the Directive, as given in Articles 5 and 6. In addition, the general objectives of the Directive, as given in Article 1, need to be taken into consideration.

3.2.5.1 Preliminary Assessment (Article 5)

Article 5 requires that Member States have representative measurements or assessments of the level of PM₁₀ for all zones and agglomerations, in time for implementation of the Daughter Directive. For most Member States, existing measurement networks for particulate material, whether TSP, PM₁₀ or black smoke, will provide at least some of the data required for this preliminary assessment. Tables 3.1 and 3.2 indicate that most Member States currently undertake some form of particle monitoring. Clearly, from these tables, most countries already have a number of monitoring stations for particulate material, but each country will need to assess whether these are sufficient to fulfil the requirement of Article 5. If not, additional representative measurements, surveys or assessments will be required.

3.2.5.2 Monitoring Requirements (Article 6)

Article 6 of the Directive sets out the requirements for assessment of ambient air quality. Four types of zone are to be considered, with different minimum assessment techniques required, as summarised in Table 3.3: Types of Zones and Assessment Techniques as defined in the Framework Directive.

Table 3.3: Types of Zones and Assessment Techniques as defined in the Framework Directive

Type of Zone	Assessment Technique
Zones above the limit value	Measurement mandatory
Zones above x% of the limit value	Measurement mandatory
Zones below x% but above y% of limit	Combination of measurements and modelling may be used
Zones below y% of the limit	Modelling or objective estimation, alone, shall be possible

After considerable discussion, the Working Group decided that x and y should be set as percentages of the limit value concentration, to take account of the typical year-to-year variation of measured PM_{10} concentrations. Examination of available data from Germany, The Netherlands and the UK indicated that interyear variability, corrected for long term trends, with a 95% confidence range (i.e. twice the standard deviation) was about 30% or less, for the annual average. Hence, a value of 70% for x related to the annual mean limit value was agreed. By similar analysis, a value of 60% for x related to high percentiles of daily values was agreed.

An extension of this approach, using three times the standard deviation gave values of y as 50% for the annual average and 40% for high percentiles of daily averages, assuming that the methods of assessment used had an uncertainty of not more than 100%.

For both x and y , the Group recognised that data from three north-western European countries might not be representative of the whole of the union, so other countries are encouraged to analyse their own data to check if the values of x and y chosen are appropriate for their situation.

The values chosen by the Working Group are summarised in Table 3.4: Proposed values for “ x ” and “ y ” below.

Table 3.4: Proposed values for “ x ” and “ y ”

	Annual Average	High Percentiles of Daily Average Values
x	70	60
y	50	40

3.2.5.3 *Number of Monitoring Stations*

The Commission’s Network Design Report states that the overall number of monitoring stations will depend on:

- ambient concentrations, in relation to limit and alert values;
- the population likely to be exposed to such concentrations;
- the scale of environmental effects (though as only human-health limit values are proposed for particles, this is not relevant here);
- the spatial variability of the pollutant.

For PM_{10} it is considered that four broad categories of monitoring site should be covered:

- urban background
- road side
- background

- industrial

Within each of these categories the monitoring stations should be located to monitor in freely mixed air where the concentrations of PM₁₀ and public exposure is expected to be highest. These sites should be positioned where the potential for human exposure, over the time scale of the limit values (i.e. daily and/or yearly), is greatest.

The first assessment of concentrations in excess of the limit value will be obtained from the Preliminary Assessment (Article 5). The Directive requirement to monitor in all agglomerations, along with the Member States' zone definition, will determine the population likely to be exposed to concentrations above the limit value. At present, the environmental effects of particulate material are not being considered and only a limit value for health protection has been proposed.

The Commission, in collaboration with the JRC and the EEA will prepare a guidance report for the further assessment of air quality, including particle concentrations in zones. The number of monitoring stations required in any zone will depend on whether this further assessment has been carried out. In zones with no preliminary assessment, the number of stations required will be defined as N_i and, in zones where an assessment has been carried out, the number of monitoring stations may approach a lower number N_o.

The approach taken by the Particles Working Group is that, in general in an agglomeration, the distance from a point measurement varies with the square root of the population. Using this square root approach avoids the problem of excessively high site numbers being required in large agglomerations. Elevated levels at roadside sites, compared with urban background sites, tend to be determined by:

- the traffic in a street (number of cars, average velocity, composition);
- the road characteristics (e.g. street canyon [76]);
- the distance from the kerbside.

The location of a street (within a town or between towns) tends to be less important than the amount of traffic and the 'canyon' character of the street. Differences in concentration due to meteorological differences between regions are mainly accounted for by differences in windspeed. As a consequence it is considered to be more important to characterise the different type of streets and traffic composition in a country than the variation between cities. It is recommended to determine the number of roadside sites per country, using the formula given below. In the assessment procedure, compliance is determined by selecting the appropriate increment to be applied for each street type, taking into account traffic flow, and then adding this to the measured cities' background concentration. In cities a complete overview of streets not in compliance with the limit value in an agglomeration may be reported.

The following formulae were therefore recommended for the determination of N_i:

Urban background sites per agglomeration	$2 \times \sqrt{\frac{I}{0.25}}, \text{ which is mathematically equivalent to } 4\sqrt{I}$
Roadside sites per country	$3 + \sqrt{P}$

Background sites per country	$\frac{A}{50000}$
Industrial sites per country	“sufficient to cover significant industrial sources”

Where:

I = the number of inhabitants of the agglomeration (millions)

P = the population of the country (millions)

A = the area of the country (square kilometres)

Fractions are to be rounded up and background sites are to be distributed according to population distribution outside of agglomerations (as defined by the Directive). This ensures coverage of agglomerations under 250000 population as well as rural and sites where secondary aerosol is important.

For N_o , it was agreed that this must be a minimum of 1 per agglomeration or zone.

Monitoring at these stations must be undertaken by a method shown to be equivalent to the reference method and which give data with the appropriate time resolution to check compliance with the limit value. Where appropriate, monitoring can also be undertaken by the random sampling method (see Section 3.2.4).

In order to facilitate checking of compliance with an alert value, should one be set, some stations will need to provide real time automatic measurements which are checked sufficiently frequently to allow initiation of the appropriate alert procedure. The number of stations needed for alert monitoring will depend on the geographical scale of the alert warning, e.g., zone, regional, country.

In zones where measurement is not mandatory, monitoring methods may not need to comply with the requirement for formal equivalence or provide data at the temporal resolution required to check for direct compliance with the limit value. However, a relationship between the limit value time average and the measured concentration needs to be established in order to use these data to estimate compliance. Moreover, the measurements could be used to validate models in the local situation, which can then be used to assess compliance.

3.2.5.4 Local Station Siting Requirements.

Once the type of location has been determined, selection of actual sites must take into account a number of factors, for example accessibility and security against vandalism (operational aspects), and provision of infrastructure (e.g. electricity, telephone).

In addition, the monitoring site should be representative of the area directly surrounding the station. As a minimum the following guidelines should be met:

- The flow around the inlet sampling probe should be unrestricted without any obstructions affecting the air flow in the vicinity of the sampler (e.g. some meters away from buildings, balconies, trees, etc.).
- In general, the inlet sampling point should be between 1.5m (the breathing zone) and 4m above the ground. A height of 1.5m would be preferred for assessment of potential human exposure near heavy traffic situations, but for practical reasons (for example, prevention of

vandalism) a height of around 2.5m is generally considered to be acceptable. For city background situations a maximum height of 4m is generally recommended, but the specific siting must be considered and higher positions (up to 8m) may be necessary in some circumstances.

- The inlet probe should not be positioned in the very near vicinity of sources in order to avoid drifting air pollution plumes (e.g. not near chimneys serving the stations own heating system).
- The samplers exhaust outlet should be positioned so that recirculation of exhaust air to the sample inlet is avoided.

The site selection procedures should be fully documented at the classification stage and compass point photographs of the surrounding area should be provided together with a detailed map. This will allow sites to be characterised in terms of local sources, topography etc. Sites should be reviewed at regular intervals with repeated documentation (including photographs) to ensure that selection criteria remain valid over time.

3.2.6 QA/QC Requirements

The contents of this section are closely based on the WHO/UNEP Report - GEMS/AIR Methodology Review Handbook Series, Volume 3, 'Measurement of Particulate Matter in Ambient Air' [56]. In addition, further information on QA/QC procedures for air quality monitoring are provided in the EC Report 'Monitoring Network Design for Air Quality Assessment' [46] the WHO UNEP GEMS/AIR Methodology Review Handbook Series, Volume 1 'Quality Assurance in Urban Air Quality Monitoring' [57] and in the EC 'Instruction Manual for Air Pollution Monitoring, Volume II: Black Smoke Monitoring' [54].

Quality assurance is a system of activities that assure that a measurement meets defined standards of quality with a stated level of confidence. A quality assurance programme includes not only standard quality control procedures (QC). i.e. the procedures undertaken to ensure a certain accuracy and precision of measurement, it also covers such topics as the definition of monitoring objectives, network design, management structure, instrument selection, staff training and procedural auditing.

High levels of quality assurance are the best means of ensuring the data within and between different networks are harmonised, i.e. that they are comparable and compatible. The quality assurance programme must ensure that the variation in the results from different types of instruments and different instruments of the same type are in an allowable range, that interlaboratory variations are minimised, and that the same variable is being measured with the same accuracy and precision across the network.

In general, laboratories responsible for air quality measurements in the Member States should seek to be accredited to formal Quality Standards. The more technically based recognised quality standards such as EN 45000 and Good Laboratory Practice are more appropriate for technical operations such as air quality monitoring, rather than more general quality systems such as EN ISO 9000. For formal accreditation, a system of controlled quality manuals, staff training and authorisation and documented "traceability chain" will be essential. The operation of this system will need to be formally assessed, on a regular basis, by external auditors.

In the discussion below, aspects of QA/QC specifically relevant to the measurement of particles are discussed.

The main operational features of a quality assurance programme will be:

- data quality objectives (DQOs)
- network design
- site selection
- monitoring station design
- equipment evaluation and selection
- site infrastructure and routine operation
- maintenance and calibration of equipment
- data review and evaluation

Network design and site selection are discussed separately in Section 3.2.5. The other aspects of quality assurance programme are considered below.

3.2.6.1 Data Quality Objectives (DQOs)

The first step in preparing any quality assurance plan is to define closely the overall monitoring objectives. These will then allow the Data Quality Objectives (DQOs) to be determined and the necessary quality assurance and quality control (QA/QC) measures to be defined. The minimum requirement will be to fulfil the requirements of the Framework Directive and, for particle measurement, additional requirements specified in the relevant Daughter Directive. The acceptability criteria for the repeatability of the PM₁₀ sampling heads and the comparability of these heads with reference samplers will be given in the CEN standard (see section 3.2.3).

When whole instruments are tested, rather than just the sampling head, a additional degree of uncertainty may be introduced, so it may be necessary to relax these criteria. In addition, routine operation of instruments on network sites, even within a documented quality system, as opposed to operation within a short duration test and intercomparison will also lead to additional uncertainty.

3.2.6.2 Monitoring Station Design

Manual particle sampler such as High Volume Samplers (HVS) are usually placed in the open air, whereas automatic samplers such as β -absorption monitors and TEOM (and BS samplers) are generally placed inside special shelters, and sample the air from outside through a sampling tube. Air conditioning may be required in the shelters, although in some cases a simple ventilation system will suffice. Water condensation and observable deposition must not occur in the sample line.

The monitoring stations should be set up and operated so as to yield representative and comparable air pollution data. To this end, the air intake system should be freely exposed to winds from all directions. The inlet should therefore be at least 1m from any vertical surface (e.g. wall) and about 1.5m from the ground, for stand-alone samplers, and about 1m from any horizontal surface (e.g. hut roof) for samplers installed within a hut or container (see also 3.2.5.4 for siting requirements).

3.2.6.3 Equipment Evaluation and Selection

Only instruments that have demonstrated equivalence to one of the reference samplers (Section 3.2.3) may be used for monitoring in zones where measurement is mandatory. Only where measurements are not mandatory, can other methods be used.

New methodologies must undergo equivalence testing before they can be used for determining compliance with the Directive. The results of validation tests for new methods should be published in a peer reviewed scientific journal.

3.2.6.4 Site Infrastructure and Routine Operation

The sampling configuration must be designed such that the concentration and distribution of particulate matter which is measured is truly representative of the concentration and distribution of particles at the sampling point (inlet).

Various operations are essential to maximise data integrity and capture rates. In general, a site visit schedule should be drawn up which makes provision for all sites to be visited regularly at specified intervals. A comprehensive calibration record and instrument checklist should be completed after each site visit and retained for subsequent QA/QC auditing. Site operators must be fully trained in all site operational procedures.

It is important that all operational procedures are fully documented and that documentation is 'backed up'. QC procedures should include statements, amongst others, on sampling method in use (type of instrument, filter paper, etc.), flow control procedures, analysis method, calibration procedures (blank and span), error handling procedures, maintenance procedures, sampling and reading requirements, reporting procedures and a report on parallel measurements with the chosen reference method [51, 54].

For manual methods, careful attention should be paid to the proper identification and labelling of all field specimens.

3.2.6.5 Maintenance and Calibration of Equipment

The importance of proper maintenance procedures cannot be overemphasised. It is only through proper instrument support that equipment can be relied upon to operate satisfactorily and for extended periods in the field. Maintenance schedules for the replacement of consumable parts, diagnostic checks and equipment overhaul should in all cases follow manufacturers recommendations.

Proper calibration of equipment is essential for obtaining accurate and reproducible air quality data. The following points relate specifically to particle samplers. It is impossible to calibrate a particle sampler by passing a known ambient particle mixture into the inlet and comparing to a measured value. Thus, QA/QC procedures focus on flow rate measurement and intercomparability. Flow rate audits should be conducted 2-4 times per year.

The mass measurement process also needs to be checked. For instance, automatic analyses should be checked with foils (β) and preweighed filters (β and TEOM). In addition, the whole measurement process of continuously operating instruments (including sampling heads and sampling lines) are to be regularly checked on site by comparison with one of the reference samplers (section 3.2.3). For manual methods, QA/QC procedures need to be established for the laboratory analysis of filters. For example, laboratory balances should be checked with standard weights and filter conditioning parameters should be properly observed.

For black smoke measurement, reflectometers need to be calibrated and operated strictly in accordance with the ISO standard and the EC Instruction Manual [54].

Periodic co-location of samplers may also be useful as a method of evaluating instrument performance and participation in international intercomparison exercises [54, 58] is also strongly recommended.

3.2.6.6 *Data Review and Validation*

Data should be reviewed by skilled personnel for identification of suspect values. This procedure can help in identifying equipment malfunction or other problems requiring attention. To ensure data integrity, it is recommended that all monitoring procedures be independently audited on a regular basis.

3.3 ASSESSMENT OF FINE PARTICULATES

3.3.1 *Introduction*

Dispersion models used for assessments with regard to regulatory purposes need to be validated against a reference situation to assure their quality. For example, a dispersion model for calculating city background levels can be validated only in cities with a well defined measurement network. If the results are satisfactory the dispersion model can be applied with caution to other, similar, cities. Measurement networks in these cities can be directed to support (giving backup evidence about the quality of the calculations) assessments by model calculations instead of serving as an independent reference situation.

3.3.2 *Assessment requirements*

Before starting an assessment, the first task is to define clearly the overall objectives of the program.

In order to fulfil the Directive's aim to "assess the ambient air quality in Member States on the basis of common methods and criteria" (Article 1), the Commission has proposed to start an European Ambient Air Quality Directive Network (EAAQDN) [59]. In addition to this, other assessment techniques are likely to be needed to meet all of the Directive's requirements [46]. The EU requirements are clearly stated in the Framework Directive. The legal requirements of the Directive for assessment¹ of ambient air quality are given in different Articles (4.3(b), 5, 6.1, 6.3, 6.4, 11.1, annex iv). They require the following 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;
- to establish criteria for spatial resolution for modelling and objective assessment methods;
- a combination of monitoring, measurements and modelling techniques may be used to assess ambient air quality where the levels over a representative period are below a level lower than the limit value;
- where the levels are below a level to be determined in the provisions referred to in Article 4(5), the sole use of modelling or objective estimation techniques for assessing levels shall be possible;
- estimate of the polluted area (km²) and of the population exposed to the pollution;

¹ Article 2 definitions: "Assessment" means any method used to measure, calculate, predict or estimate the level of a pollutant in the ambient air.

- details of those factors responsible for the excess;
- estimates of the improvement of air quality planned and of the expected time required to attain these quality objectives.

In the paragraphs following the above considerations have been worked out for particulates, under the assumption that both a value for a yearly average and a maximum daily/high percentile value will be recommended and that particulates are not relevant for eco-systems.

3.3.3 *Assessment models for particulates*

3.3.3.1 *Long term (yearly average)*

For calculation of yearly average concentrations a series of models are available, from local scale (dispersion around an industrial site, a line source e.g. traffic in a street over a city) to European scale [60, 61].

The uncertainty of the calculations, under the assumption that there are no errors in the emission database, vary typically between 10 and 50 % for models used for regulatory purposes (4th Workshop on Harmonisation within Atmospheric Dispersion Modelling for Regulatory Purposes, 1996).

The main source of error, however, is the quality of the emission inventories. Especially for PM₁₀ the uncertainty in the emissions is rather high (in the range 20-70%) preventing high quality calculations. As errors in the calculated air pollution load are proportional to those of emissions, improvement of emission inventories is necessary to gain satisfactory model results.

For the time being, the range of uncertainty of a certain dispersion model and a given emission data base has to be determined from comparison with monitoring data in those areas where results of measurements are available. If such validation cannot demonstrate that the range of uncertainty of the dispersion model combined with the emission data base is below x or y % of the limit value (see Table 3.3 and 3.4), this model cannot be used for compliance checking.

3.3.3.2 *Short term (percentiles)*

A concept similar to that outlined above for chapter 3.3.3.1 is recommended. Model uncertainties of 24-hour-mean-values amount up to factor of 2, i.e. 200% (Borrego, Portugal, 4th Workshop on Harmonisation). This is comparable to 99-percentile errors. Concerning 98-percentiles of daily means lower errors may be expected. An intercomparison of regulatory models has shown that the position of maximum 98-percentiles may vary by several km (Mensink, Belgium, 4th Workshop on Harmonisation).

3.3.4 *Other Assessment Techniques*

In addition to dispersion models, concentrations of PM₁₀ can also be estimated using other associated pollutants as indicators. The use of such indicators is appropriate in situations where the relationship between the relative proportions of PM₁₀ concentrations and other pollutants is robust and well characterised. Under these conditions, for example, PM₁₀ concentrations may be predicted at roadside sites using measured concentrations of other associated vehicular generated pollutant such as NO_x, CO or black smoke. The technique is not appropriate in locations where concentrations of PM₁₀ and other pollutants are dominated by industrial point sources with unpredictable or sporadic emissions.

3.3.5 QA/QC for assessments

There is a clear need for reliable information documenting the quality of ambient air, and the response to control measures, in urban areas. Such information provides the basis for decision-making and the development of planning strategies. In order to be sure that decisions are properly founded, it is necessary to be certain that the recorded measurements or model calculations genuinely reflect the existing situation, in other words the data must be of clearly defined and documented quality.

Quality assurance and control (QA/QC) is thus an essential feature of any successful environmental assessment program. For dispersion models some guidelines are available [59 - 61].

3.4 DATA PRESENTATION AND EVALUATION

3.4.1 Introduction

A final, validated, air quality dataset is - in itself - of limited utility. To be of use for policy development, air pollution management, effects investigations or scientific research, the data must be collated, assessed and disseminated. A minimum level of data assessment, is the production of annual (and preferably monthly) data summaries. These may use simple statistical and graphical analysis methods. Suitable report formats for this purpose can be readily designed (see for instance [64]). More regular data reporting, even daily or hourly, may be appropriate for some network objectives (e.g. smog-alert systems), although in such cases it must be made clear that the disseminated data cannot be fully validated or quality assured.

Dissemination of provisional or validated data only represents a first step in meeting assessment requirements. The information should provide a background for decision makers, as well as a basis for developing and testing scientific hypotheses and models of processes related to air quality and its impact on man and his environment. In order to make the fullest use of ambient air quality measurements, it is necessary to have corresponding and compatible high quality data on a variety of other socio-economic and geographic factors in the data analysis. For example, using the data to gain insight into the atmospheric processes leading to elevated pollutant concentrations requires access to emission and meteorological information; assessing resultant health and ecosystem impacts requires information on population distributions, epidemiology, social factors and health effects.

Monitoring and emissions data are generally intended for use in developing strategies for the reduction of effects, i.e. for management of air pollution at a city or national level. The strength of a European monitoring database such as APIS/EoI is that it can be used for long-term assessment of the impact of control measures, and to estimate the situation in cities/countries for which only limited data are available by comparison with similar 'representative' cities.

The specific requirements of the Directive Article 11, Transmission of Information and Reports, must also be addressed.

3.4.2 Presentation of Measurements

With respect to measurements, Article 11 requires that the Commission be informed "of occurrence of levels exceeding the limit value [plus a margin of tolerance], the date(s) or period(s) when such levels were observed and the values recorded in the nine-month period after the end of the year." In addition, the reasons for such exceedences must be given, together with details of plans to be implemented to attain compliance and, details of progress on these plans (every 3-years). In order to harmonise the provision of

data, it is suggested that the Commission prepare a simple proforma or protocol for the supply of this information, possibly along the lines of that used currently for the Directive 80/779/EEC.

It is important to note that, where percentile concentrations are required then the Commission should specify the method of calculation to be used. Currently, the method specified in the Directive 85/203/EEC (NO₂ Directive) and Directive 92/72/EEC (O₃ Directive) differs from that recommended by the Commission for percentile calculations in the Directive 80/779/EEC (SO₂/SMP Directive). These methods were investigated by Beier [65]. From the ordered set of N values, the p th percentile is taken as the value of the component of rank k where k is calculated as follows:

for 85/203/EEC and 92/72/EEC

$$k = \text{INT}(q * N) \quad \text{i.e. rounded to the nearest whole number}$$

but, for Directive 80/779/EEC

$$k = \text{INT}(q * N) + 1 \quad \text{i.e. rounded to the next higher whole number.}$$

The latter formula yields more safe results in case of small sample sizes N . Therefore, this formula is recommended for the calculation of percentiles.

Generally, these methods lead only to small differences in calculated statistics, but this could be significant if extreme percentiles, such as 99.5th, are required.

Hourly and daily values will also have to be submitted to the European Environment Agency (EEA) under the terms of the Exchange of Information Decision (28/96/EC of 26 February 1996). In its current form, site information is held in the GIRAFE database and corresponding measurement values are held in the related APIS database. Data held in these databases have recently been made available on CD-ROM by the EEA-Topic Centre on Air Quality (EEA-TC). The EEA-TC proposes to improve the data structure of these databases and enable access via the Internet. The new database will be called AIRBASE and formats for data exchange are currently being developed.

3.5 SUMMARY OF KEY POINTS AND RECOMMENDATIONS

The Directive requires both monitoring (measurement) and assessment of air quality.

3.5.1 *Measurement*

Commercial manual and automatic instruments are available for monitoring TSP, PM₁₀ and other particle size fractions in the timescale appropriate for checking compliance with the limit value.

Monitoring must be undertaken by a method shown to be equivalent to the reference sampler or to one of the two transfer reference samplers and which gives data with the appropriate time resolution to check compliance with the limit value.

The CEN standard methodology should be used to check samplers against reference sampler or equivalent reference samplers.

In areas where monitoring is required, data capture over the year should be at least 75% for automatic methods and 90% of planned measurements for manual methods. Lower data capture may be acceptable in other areas.

Values for x and y (Article 6), defining zones for monitoring should be set at:

x: 70% for annual average 60% for high percentiles of daily averages

y: 50% “ 40% “

The number of monitoring stations per zone is defined as:

N_o in areas where assessment has been carried out

N_i in other areas

N_o is a minimum number of stations, i.e. 1

Values for N_i are defined as follows:

Urban background sites per agglomeration	$2 \times \sqrt{\frac{I}{0.25}} = 4\sqrt{I}$
--	--

Roadside sites per country	$3 + \sqrt{P}$
----------------------------	----------------

Background sites per country	$\frac{A}{50000}$
------------------------------	-------------------

Industrial sites per country	“sufficient to cover significant industrial sources”
------------------------------	--

Where:

I = the number of inhabitants of the agglomeration(millions)

P = the population of the country (millions)

A = the area of the country (square kilometres)

Where possible, laboratories undertaking monitoring should seek formal accreditation under EN45000 or Good Laboratory Practice, rather than more general quality standards such as EN ISO 9000.

Quality control procedures should be fully documented and cover all aspects of the measurement process.

3.5.2 Assessment

Dispersion models used for assessments with regard to regulatory purposes need to be carefully validated against a reference situation to assure and control their quality. In addition to dispersion models, concentrations of PM_{10} can also be estimated using other pollutants as indicators, although this technique is not appropriate in locations where concentrations of PM_{10} and other pollutants are dominated by industrial pollutant sources with unpredictable or sporadic emissions. In order to be sure that decisions are properly founded, it is necessary to be certain that the recorded measurements or model calculations genuinely reflect the existing situation; in other words, the data must be of clearly defined and documented quality.

3.5.3 Data Presentation

Monitoring data showing levels, date, period and location of any exceedence of the limit value must be communicated to the Commission. It is suggested that a pro forma or protocol is established to achieve this in a harmonised way

To calculate percentile concentrations it is recommended to use the method given in Directive 80/779/EEC (SO₂/SPM Directive).

Full annual datasets will be provided to the EEA under the terms of the Exchange of Information Decision.

4. COSTS

4.1 INTRODUCTION

The sparse PM₁₀ results make it difficult to establish a representative overview of PM₁₀ concentrations and trends in the member states. The results presented in Chapter 1 suggest, however, that the present values in the majority of the member states exceed the recommended limit values. Abatement strategies have, therefore, to be developed if the currently planned precautions do not reduce emissions sufficiently. These must include a definition of the needs considering a weighing of the risks and costs. The present concentrations, as summarized in tables A2, show a predominantly south to north gradient. In Spain and probably also the other Mediterranean countries, a reduction of the total emissions by a factor two or more may be necessary to achieve compliance with the limit values recommended (section 2.7.4) and the natural particulate input in these areas may make it difficult to comply with the standard, while major emission reductions in the northern part of Europe are likely to be required if the limit values described in section 2.7.4 are implemented. A fundamental problem at this stage is to establish a representative overview of the present situation based on the relatively sparse long term PM₁₀ measurements. A better estimate may be made by more detailed model calculations taking dispersion, deposition and atmospheric chemistry into account. Further, the more comprehensive collection of results for total suspended particulate matter should be considered.

The proposed limit values for particulate matter are based on PM₁₀ concentrations, which are only a fraction of the total aerosol in the atmosphere. The already implemented or proposed emission abatement strategies do not include specification of the particle size distribution of the particulate matter. The research into the health effects of SPM has to a large extent been based on PM₁₀. However, recent studies (cf. Chapter 2) indicates the fine particles are potentially of major importance and size fractions such as PM_{2.5} might give a more well defined measure of the damaging effect of the particulates. As stated in Chapter 2, there may be good reasons to consider fractions other than PM₁₀ and that the limit values set for PM₁₀ might have to undergo revisions at a later stage. In order to anticipate this development the calculated reduction scenarios should include assessment of their efficiency as function of particle size.

Currently, planned actions should be taken into consideration. There are few international initiatives that directly aim at a reduction of particles emission. Of greatest importance seems to be the emission regulations for diesel cars. The SO_x and NO_x Protocols within the UNECE require an extensive reduction of these species based on the needs for a decrease of deposition on ecosystems. The emission reductions agreed in that form will also reduce the formation of secondary particles.

In order to achieve the necessary reductions, abatement strategies may depend on regional differences and scale. For industrial areas regulation against point sources may be sufficient, while a combined strategy for several source types e.g. traffic, industry and resuspended material may be necessary at other places. An important issue will be the extent to which abatement strategies will effect episodic compared with long term average concentrations.

Particle emissions are different from those of the other pollutants included in the first phase of the EU framework Directive in that a great number of source types contribute to the total ambient concentrations. Even if anthropogenic sources are dominating in the major part of the EU area, natural sources may in some areas contribute significantly to the integrated particle level on a local or regional scale.

4.2 ABATEMENT TECHNIQUES

A summary of the main sources of PM₁₀ and possible abatement techniques is given in Table 4.1.

Table 4.1 Overview of PM₁₀ sources and possibilities for reduction.

Source type	Possible abatement	Effected by already initialized abatement	Comments
Anthropogenic sources			
Road transport - direct emission	Improved combustion, fuel, filtering	EU regulation on diesel emission	Cost estimates in Auto-Oil program
- resuspension (including sanding of roads)	Road consolidation Better sanding practices Improved road maintenance and sanitation		
Domestic heating (mainly wood and coalburning)	Filters or catalysts on stoves and furnaces		
Other combustion (industrial, waste, power) - primary	Filtering, better combustion	Application of BAT ¹⁾	Information on cost from industries through CONCAWE
- secondary from NO _x , SO _x photochemical	Fluegas cleaning VOC and NO _x reduction	UNECE NO _x , SO _x protocols	
Agriculture livestock, poultry	Removal of particles from ventilation exhausts. Reduction of ammonia emissions.		
Forest, heathland and Agricultural fires	Avoid stubble burning. Forest fire prevention		Already implemented in many countries
Natural sources			
Sea spray			Of minor importance for PM ₁₀ concentration
Soil resuspension	Plant coverage of unused fields	Prevention of eutrophication of freshwater and near coast sea water	Especially important in the Mediterranean basin
Long range dust transport and volcano emissions			Important in the Mediterranean basin

¹⁾ Best available technology.

The most important PM₁₀ sources are described in Chapter 1. It is not possible to denote one or even a couple as the most important on an EU scale. Abatement techniques may thus have to be implemented at a regional scale. Abatement policies for other pollutants may in many cases influence particles emissions, and the assessment of relative costs and benefits of control will need to take this into account.

In order to assess the relative effectiveness of different abatement strategies, it is important to know the contributions of different source types to measured concentrations of particles. This can be achieved through source-apportionment analyses of measurements, or from comprehensive emission inventories and dispersion modelling of PM₁₀. The contribution to measured levels from secondary aerosols (mainly SO_x and NO_x) should also be determined. However, most inventories from the member states are based on TSP. Specific PM₁₀ inventories are only available from a few countries (see Annex 1), together with a Dutch emission inventory for primary man-made PM₁₀ in Europe.

An important tool for studying the effect of different control strategies on the atmospheric concentrations will be dispersion models, which should include both the atmospheric chemistry for formation of secondary aerosols, and the deposition rate as a function of particle size. These models should be coupled to an exposure model in order to assess the health impact and thus make it possible to establish the proportion and likely effects of the abatement policy. The Dutch emission inventory will be combined with population maps and the WHO risk assessment are incorporated in this model. Dispersion models for particles are in a state of rapid development, but uncertainties remain, not least over emission inventories. Recent reviews are found in [60] and [61].

4.3 BENEFITS OF THE REDUCTION OF PM₁₀

The primary consideration for the proposed limit values in Chapter 2 has been the protection of human health. As a threshold for zero risk could not be established, the eventual determination of limit values will involve the consideration of what constitutes acceptable risk. The health benefits should be evaluated by considering the outcome of the WHO working group [32] incorporating the more recent PEACE and APHEA studies [39].

Reduction of PM₁₀ emissions in Europe are expected as a result of measures already planned or decided. The reduction of SO_x and NO_x is expected to have an effect on the PM₁₀ concentrations, but the actual reduction of the ambient concentrations may not be linearly related to the emission reduction because the formation rate for the secondary aerosols also depends on the concentration of oxidizing and neutralizing species (e.g. OH-radicals and ammonia) in the atmosphere. The auto-oil calculations are for total particulate matter, but almost all the particles considered are smaller than 10 µm.

Table 4.2: Expected reduction of emissions compared to 1990 for species which contribute to PM₁₀. The SO_x and NO_x vales are based on current legislation scenarios for the EU15 countries. The total particulates results are from the Auto-Oil Programme year 2000 proposals [74].

Species	PM ₁₀ source	1990 kton	2000	2010	Reduction from 1990 to 2010
NO _x	secondary	23000		15000	49 %
SO _x	secondary	38000		19000	35 %
Total particulates	road traffic	125	86	45	64 %

Information on building and materials damages from particles are collected by DGXII. Preliminary results indicate that compliance with the currently proposed range of limit values would represent a significant improvement. The soiling of plants is not known to reduce the yield of crops, but it may, together with reduced visibility due to fine aerosols in the atmosphere, be unpleasant and thus have an impact on tourism and not least the general experience of enjoyment of life.

4.4 DISCUSSION

The EU standards for fine particulates are expected to be based on PM₁₀, but a change to or inclusion of PM_{2.5} seems likely at some point in the future. It will hence be appropriate to consider to what extent the PM_{2.5} level will be influenced by proposed abatement measures for PM₁₀.

PM₁₀ differs from the other pollutants included in the first phase of the framework directive. Most of the available information concerning emissions and ambient concentrations concerns TSP. Moreover, the natural background levels are significant in many regions. The behaviour of combustion aerosols and mechanical derived aerosols (both natural and anthropogenic) may be different due to different particle size distributions. Furthermore, concentrations of particles arising from primary emissions can be supplemented by secondary aerosol particles formed primarily from emissions of SO₂, NO_x and ammonia. In order to assess the effectiveness and benefits arising from abatement strategies for particles, it is therefore necessary to estimate the contributions to ambient particle levels from the various sources of primary, secondary and naturally occurring particles. Unless such estimates are available for individual urban areas, it will be difficult to assess the effects of current policies already in place or of new ones.

In assessing costs, particularly in the context of abatement measures which control several pollutants at once, care must be taken not to assign the total costs to any one pollutant. This is particularly important in relation to measures to reduce emissions from motor vehicles. Similarly, in assessing costs, the potentially significant differences between Member States should be taken into account.

5. REFERENCES

- [1] Prospero J.M., R.A. Glaccum and R.T. Nees (1981). Atmospheric transport of soil dust from Africa to South America. *Nature* 289, 570-572.
- [2] Savoie D.L. and J.M. Prospero (1982). Particle size distribution of nitrate and sulfate in the marine atmosphere. *Geophys. Res. Letter* 9, 1207-1210.
- [3] Nicholson K.W. (1988). A review of particle resuspension. *Atmos. Environ.* 22, 2639-2651.
- [4] Rahn K.A., R.D. Borys, G.E. Shaw, L. Schütz and L. Jaenicke (1979). In: *Saharan Dust: Mobilization, Transport, Deposition*. Scope 14. Ed: C. Morales. Wiley and Sons, Chichester UK.
- [5] Haulet R., P. Zettwoog and J.C. Sabroux (1977). Sulphur dioxide discharge from Mount Etna. *Nature* 268, 715-717.
- [6] Möller D. and H. Schieferdecker (1985). A relationship between agricultural NI-13 emissions and the atmospheric SO₂ content over industrial areas. *Atmos. Environ.* 19, 695-700.
- [7] Malinconico L.L. (1979). Fluctuations in SO₂ emission during recent eruptions of Etna. *Nature* 278, 43-45.
- [8] OECD Report. *Methods of Measuring Air Pollution*. Organization for Economic Co-operation and Development. Paris 1964.
- [9] ISO 9835:1993(E) *Ambient air-Determination of a black smoke index*.
- [10] US Federal Register (1987). *Ambient monitoring reference and equivalent methods*. United States Environmental Protection Agency. Federal Register 40 CFR Part 53, 1 July 1987.
- [11] ISO-7708-1995. *Air quality - Particle size fraction definitions for health-related sampling*.
- [12] Expert Panel on Air Quality Standards. *Particles*. HMSO, London 1995.
- [13] Departments of the Environment, Health and Transport. *Health Effects of Particles*. UK November 1995.
- [14] Quality of Urban Air Review Group. *Airborne Particulate Matter in the United Kingdom*. UK, May 1996.
- [15] Pavoni J.L., J.E. Heer Jr. and D.J. Hagerty (1975). *Handbook of solid waste disposal. Materials and Energy recovery*. Van Nostrand Reinhold Company. New York 1975.
- [16] Bouscaren R., C. Veldt, K.H. Zierock and B. Lübker (1987). CORINAIR project, Programme CORINE, National Experts Meeting held in Brussels, 7-8 May 1987.
- [17] Crutzen P.J. and M.O. Andreae (1990). Biomass burning in the tropics: Impact on Atmospheric Chemistry and Biogeochemical Cycles. *Science* 250, 1669-1678.
- [18] Velez Muñoz R. (1990). Los incendios forestales en España. *Ecología*. Fuera de serie n°1. 213-223.

- [19] Blanchard D.C. and A.H. Woodcock (1980). The production, concentration and vertical distribution of the sea-salt aerosol. *Ann. New York Acad. Sci.* 338, 330-347.
- [20] Junge C.E. (1972). Our knowledge of the physico-chemistry of aerosols in the undisturbed marine environment. *J. Geophys. Res.* 77, 5183-5200.
- [21] Warneck P. (1988). *Chemistry of the Natural Atmosphere*. Internat. Geophysics Series, Vol. 41.
- [22] Löye-Pilot M.D., J.M. Martin and J. Morelli (1986). Influence of Saharan dust on the rain acidity and atmospheric input to the Mediterranean. *Nature* 321, 427-428.
- [23] McDowell W.H., C. Gines Sanchez, C. E. Asbury and C.R. Ramos Perez (1990). Influence of sea salt and long range transport on precipitation chemistry at El Verde, Puerto Rico. *Atmos. Environ.* 11, 2813-2821.
- [24] Roda F., J. Bellot, A. Avila, A. Escarra, J. Piiol and J. Terradas (1993). Saharan dust and the atmospheric inputs of elements and alkalinity to Mediterranean ecosystems. *Water, Air and Soil Poll.* 66, 277-288.
- [25] Whitby K. T. (1978). The physical characteristics of sulfur aerosols. *Atmos. Environ.* 12, 135-159.
- [26] Calvert J. G., Lazrus A., Kok G. L., Heikes B. G., Walega J. G., Lind J., and Cantrell C. A. (1985). Chemical mechanisms for acid generation in the troposphere. *Nature* 317, 27-35.
- [27] Chow J.C. (1995). Measurement methods to determine compliance with ambient air quality standards for suspended particles. *J. Air & Waste Manage. Assoc.* 45, 320-382.
- [28] Lundgren D.A. and H.J. Paulus (1975). The Mass Distribution of Large Atmospheric Particles. *J. Air Poll. Control Assoc.* 25, 1227 - 1231.
- [29] Laskus L., D. Bake, R. König, A. van der Meulen and B.G. van Elzakker (1989). Measurement of particle size distribution and dust concentration. *Staub-Reinhalt. Luft* 49, 395-400.
- [30] Lee S.D., T. Schneider, L.D. Grant and P.J. Verhoek (1986). *Aerosols Research, Risk Assessment and Control Strategies*. Lewis Publishers Inc. Chelsea, Michigan, pp. 521 - 593.
- [31] Monn Ch., O. Braendli, G. Schaeppi, C. Schindler, U. Ackermann-Liebrich, P.H. Leuenberger and SAPALDIA Team (1995). Particulate matter <10 μm (PM₁₀) and total suspended particles (TSP) in urban, rural and alpine air in Switzerland. *Atmos. Environ.* 29, 2565-2573.
- [32] WHO Regional Office for Europe: Update and Revision of the Air Quality Guidelines for Europe. Meeting of the Working Group 'Classical' Air Pollutants, Bilthoven, The Netherlands, 11-14 October 1994 (EUR/ICP/EHAZ 94 05/PBOI).
- [33] Working Paper for the final consultation of the Working Group 'Classical' Air Pollutants, Bilthoven, The Netherlands, 28-31 October 1996.
- [34] Katsouyanni K., D. Zmirou, C. Spix, J. Sunyer, J.P. Schouten, A. Pönkä, H.R. Anderson, Y. Le Moullec, B. Wojtiniak, M.A. Vigotti and L. Bacharova: Short-term effects of air pollution on

- health: a European approach using epidemiological time-series data. *Eur. Respir. J.* 8 (1995) 1030-1038.
- [35] EPA Air Quality Criteria for Particulate Matter, EPA/600/P-95/001aF, April 1996.
- [36] The APHEA project. Short term effects of air pollution on health: a European approach using epidemiological time series data. *J. Epidemiol. Commun. Health* 50 (1996) Suppl. 1.
- [37] Health Effects Institute: Particulate Air Pollution and Daily Mortality. Replication and Validation of Selected Studies. The Phase I Report of the Particle Epidemiology Evaluation Project. Health Effects Institute, Cambridge, August 1995.
- [38] US Environmental Protection Agency, Office of Air Quality Planning and Standards. Review of the National Ambient Air Quality Standards for Particulate Matter: Policy Assessment of Scientific and Technical Information. External Review Draft. April 1996.
- [39] 7th ISEE/5th ISEA conference, August 30 - September 1, 1995, Noordwijkerhout, The Netherlands: Presentations of first results from APHEA and PEACE projects and other presentations.
- [40] Proc. Colloquium on Particulate Air Pollution and Human Mortality and Morbidity, 24-25 January, 1994, Irvine, California. *Inhalation Toxicol.* 7 (1995) No. 1.
- [41] Dockery DW, Pope CA III, Xu X, Spengler JD, Ware JH, Fay ME, Ferris BG Jr, and Speizer FE (1995). An Association Between Air Pollution and Mortality in Six U.S. Cities. *N Engl J Med* 329, 1753-1759.
- [42] Pope CA III, Schwartz J, Ransom MR (1995). Particulate Air Pollution as a Predictor of Mortality in a Prospective Study of US Adults. *Am. J. Crit. Care Med.* 151, 669-674.
- [43] Seaton A., MacNee W., Donaldson K., Godden D. (1995). Particulate air pollution and acute health effects. *Lancet* 345, 176-178.
- [44] Wichmann H.E. and J. Heinrich (1995). Health Effects of High Level Exposure to Traditional Pollutants in East Germany - Review and Ongoing Research. *Environ. Health Perspect.* 103, Suppl. 2, 29-35.
- [45] US Environmental Protection Agency, Office of Air Quality Planning and Standards. Review of the National Ambient Air Quality Standards for Particulate Matter: Policy Assessment of Scientific and Technical Information. External Review Draft. November 1995.
- [46] Stevenson K.J., J. Vallance-Plews, J. Bower and G.J. Dollard (1995). Monitoring network design for air quality assessment. AEA, Report to the EC Joint Research Centre and DG XI.
- [47] RIVM/NILU (1995), Dobris background report.
- [48] Rodes, Ch.E., D.M: Holland, L.J. Purdue and K.A. Rehme (1985). A field comparison of PM₁₀ inlets at four locations. *J. Air Poll. Contr. Ass.* 35, 345-354.
- [49] Holländer, W., K.H. Zierock, B.G. van Elzakker, A. van der Meulen, L. Laskus and G. Morawitz (1988). A reference method for the measurement of total suspended particulates. Report on a study of the Commission of the European Communities and the Federal Environmental Agency, Berlin

- [50] Executive Summary on Studies of the Commission of the European Communities on the Development of a SPM Reference Method. Doc. N 13 of CEN/TC/ 264/WG 6 (1990).
- [51] Air Quality - Field test procedure to demonstrate equivalence of sampling methods for the thoracic fraction of suspended particulate matter (SPM) with a reference sampling method for the thoracic fraction of SPM. CEN Standard prEN 12341 (1996).
- [52] Annema J.A., H. Booij, J.M. Hesse, A. van der Meulen and W. Siooff. Integrated Criteria Document Fine Particulate Matter. National Institute of Public Health and the Environment, Bilthoven, The Netherlands. Report No. 601014015.
- [53] Air Quality - Evaluation of the designated CEN field test procedure to demonstrate equivalence of sampling methods for the thoracic fraction of suspended particulate matter (SPM) with a reference sampling method for the thoracic fraction of SPM. Doc. N 74 of CEN/TC 264/WG 6 (1996).
- [54] Christolis M., P. Clayton, P. Hecq, M. Payrissat and F. Petit-Coviaux. Instruction Manual for Air Pollution Monitoring. Volume 11 - Black Smoke. CEC JRC Ispra, Italy. Report EUR, 14550/11 EN.
- [55] US EPA. National Air Quality and Emissions. Trends Report (1992). Research Triangle Park, North Carolina 27711. Report 454/r-93-031
- [56] WHO/UNEP Methodology Review Handbook Series - Volume 3 - Measurement of Suspended Particulate Matter in Ambient Air (1994). UNEP, Nairobi. Report Reference WHO/EOS/94.3, UNEP/GEMS/94.A.4.
- [57] WHO/UNEP Methodology Review Handbook Series - Volume 1 - Quality Assurance in Urban Air Quality Monitoring (1994). UNEP, Nairobi. Report Reference WHO/EOS/94. 1, UNEP/GEMS/94.A.2.
- [58] Payrissat M., K.I. Stevenson and B. Lingner (1990). 2nd European Quality Assurance Programme for Sulphur Dioxide and Suspended Particulates Measurements. CEC JRC Ispra, Italy. Report EUR 12334 EN.
- [59] Cartalis C. (1996). Recommendations for an assessment approach at the European level. ETC/AQ, Draft report to EEA.
- [60] Moussiopoulos N., E. Berge, T. Bohler, F. de Leeuw, K. Gronskei, S. Mylona and M. Tombrou (1996). Models for ambient air quality and pollutant dispersion/transformation. ETC/AQ, Draft report to EEA.
- [61] Leeuw F. de, E. Berge, K. Gronskei and M. Tombrou (1996). Review on requirements for models and model applications. ETC/AQ, Draft report to EEA.
- [64] Bower J. S., Broughton G. F. J., Willis P. G. and Clark H. (1995). Air Pollution in the UK 1993/94. National Environmental Technology Centre, AEA, Culham,
- [65] Beier R. (1988). General Description of Statistical Treatments of SO₂, SPM and NO₂ Measurement Data in the EEC. Presented Internat. Workshop on Harmonisation of the Technical Implementation of EEC Air Quality Directives. November 22-24, 1988, Lyon, France.

- [66] WHO European Centre for Environment and Health (1995). Concern for Europe's Tomorrow: Health and the Environment in the WHO European Region. Wissenschaftliche Verlagsgesellschaft, Stuttgart, 1995.
- [67] Statistisches Bundesamt: Statistisches Jahrbuch 1995 für die Bundesrepublik Deutschland. Verlag Metzler-Poeschel, Stuttgart 1995.
- [68] Gamble J., King D., Stephenson R., Tordoir W.F., Simpson B.J.: Air quality standard for particulate matter. CONCAWE report no. 95/62. CONCAWE, Brussels, May 1996.
- [69] Thurston, G.D. (1996). A Critical Review of PM₁₀-Mortality Time-Series Studies. J. Expos. Anal. Environ. Epidemiol. 6, 3-21.
- [70] Ito, K. and G.D. Thurston. (1996). Daily PM₁₀/Mortality Associations: An Investigation of At-Risk Subpopulations. J. Expos. Anal. Environ. Epidemiol. 6, 79-95.
- [71] Lipfert FW, Wyzga RE (1995). Uncertainties in Identifying Responsible Pollutants in Observational Epidemiology Studies. Inhalation Toxicology 7: 671-689.
- [72] McClellan, R.O. (1989). Reflections on the Symposium: Susceptibility to Inhaled Pollutants. In: Utell, M.J. and R. Frank (eds.) Susceptibility to Inhaled Pollutants, ASTM STP 1024, Am. Soc. Test. Materials, Philadelphia, 1989, pp. 224-231.
- [73] European Commission (1996). Air Quality Report of the Auto-Oil Programme Report of Subgroup 2.
- [74] European Commission (1996). Communication to the Council and the Parliament on the future strategy for the control of atmosphere emissions from road transport taking into account the results from the Auto-Oil programme
- [75] United States Environmental Protection Agency (April 1995). AP42 Emission Factors. Superintendent of Documents, US Government Printing Office, Washington DC, USA
- [76] Boeft J. den, Eerens H. C., Tonkelaar W. A. M. den, Zandveld P. Y. J. (1996). CAR-International: a simple model to determine city street air quality, Sci. Total Environ., vol 189/90, 321-326
- [77] Pandis S. N., Paulson S. E., Seinfeld J. H. and Flagan R. C. (1991). Aerosol formation in the photooxidation of isoprene and B-pinene. Atmos. Environ. 25A, 997-1008.
- [77] Wordley J., Walters S. E. and Ayres J. G. (1997). Short term variations in hospital admissions and mortality and particulate air pollution. Occupational and Env. Medicine, 54, 108-116.

6. ANNEX 1: PARTICULATE MATTER EMISSIONS INVENTORIES

Table A1.1 Particulate Matter Emissions and Major Sources in Germany

Source	Emission (kt)				
	1990	1991	1992	1993	1994
Loading/unloading of bulk goods	281	260	200	198	193
Industrial processes	431	151	130	130	130
Traffic	104	105	110	115	115
Domestic coal burning	134	90	64	63	55
Small consumers	185	84	55	62	59
Industrial combustion processes	447	186	102	86	81
Power and heating plants	477	317	203	182	173
TOTAL	2059	1193	864	837	805

Table A1.2 Estimated UK PM₁₀ Emissions

	Source	1970	1980	1990	1991	1992	1993	1993 as %
1	Public power etc.	41	58	39	39	39	40	15 %
	Coal	38	57	38	39	38	38	14 %
	Fuel Oil	3	1	1	1	1	1	0 %
	Other	0	0	0	0	0	0	0%
2	Commercial Institutional & Residential Combustion Plants	226	96	45	47	45	42	16 %
	Domestic	210	88	39	41	37	37	14 %
	Other	26	9	6	5	5	5	2 %
3	Industrial Combustion Plants & Processes with Combustion	131	60	46	45	44	44	17 %
	Refineries	10	10	6	6	6	7	3 %
	Iron and Steel	56	27	21	21	20	20	8 %
	Other	65	33	19	19	18	18	7 %
4	Non-Combustion Processes	63	63	63	63	63	63	24 %
	Construction	4	4	4	4	4	4	2 %
	Industrial Processes	30	30	30	30	30	30	11 %
	Mining and Quarrying	29	29	29	29	29	20	11 %
5	Extraction & Distribution of Fossil Fuels	0	0	0	0	0	0	0
6	Solvent Use	0	0	0	0	0	0	0
7	Road Transport	44	53	68	69	68	66	25 %
	Petrol Exhaust	11	15	17	16	14	13	5 %
	Diesel Exhaust	32	35	47	49	50	49	19 %
	Non-Exhaust (Tyres and Brakes)	2.3	3	4.4	4.4	4.4	4.4	2 %
8	Other Transport	20	9	7	7	7	7	3 %
9	Waste Treatment and Disposal	0	0	0	0	0	0	0
10	Agriculture	2	1	1	1	1	1	0
11	Nature	0	0	0	0	0	0	0
	Total	527	340	269	271	267	263	100 %

Data taken from the Third Report of the UK Quality of Urban Air Review Group, 'Airborne Particulate Matter in the UK', May 1996 [14]

Table A1.3 Emissions of PM₁₀ in Greater London, UK, in 1990

Source	Emissions (tonnes)	Emissions as %
Cars	1430	16
Taxis	182	2
LGV	699	8
MGV	2508	28
HGV	1968	22
Buses	766	9
Motorcycles	75	1
Trains	16	0
Water	5	0
Air	558	6
Domestic	12	0
Industrial	212	2
Commercial and Institutional	325	4
Construction	220	2
Total	8975	

Data taken from the Third Report of the UK Quality of Urban Air Review Group, 'Airborne Particulate Matter in the UK', May 1996 [14]

7. ANNEX 2: PARTICLE CONCENTRATION DATA FROM MEMBER STATES

Table A2.1: Finland(PM₁₀)

Site Name	Site Type	Pollutant	Annual Mean μgm^{-3}			98th %ile of daily values μgm^{-3}			Maximum of daily values μgm^{-3}		
			1992	1993	1994	1992	1993	1994	1992	1993	1994
ESPOO - LEPPÄVAARA	UB	PM ₁₀		22	22		53			56	70
HELSINKI - KAISANIEMI	UB	PM ₁₀	25	23					60	51	
HELSINKI - VAILLIA 2	UB	PM ₁₀	23	22					65	62	
HELSINKI - TÖÖLÖ	UT	PM ₁₀		26	28					82	150
JYVÄSKYLÄ - LYSEO	UT	PM ₁₀	17	17	14	63	50	46	99	69	96
RAISIO - RAISION KAUPUNGINATALO	UT	PM ₁₀		19						87	
LAHTI - TORI	UT	PM ₁₀	17						45		
LAHTI - MÖYSÄ	UT	PM ₁₀	13						45		
OULU - KESKUSTA	UT	PM ₁₀	24	25		88	89		204	119	
OULU - PYYKÖSJÄRVI	Ind.	PM ₁₀	14	14		38	32		67	54	
TAMPERE - KESKUSTORI, KIOSKI	UT	PM ₁₀		25						90	
VALKEAKOSKI - TIETOLANHARJU	UT	PM ₁₀		28						228	
KOTKA - KARHULAN KESKUSTA	UT	PM ₁₀	27						137		

VANTAA - TIKKURILA ASERNATIA	UT	PM ₁₀			24						70
KOKKOLA - PIHIAJA	Ind.	PM ₁₀		16			42			46	
LOHJAN KUNTA - VIRKKALAN	Ind.	PM ₁₀	19		18				70		
PORI - VAINOJA	Ind.	PM ₁₀		19			56			101	
RAAHE - VARIKKO	Ind.	PM ₁₀		16	16		40	47		56	222

QA/QC Information

HIVOL Sampler - yearly check of flow determination (information is not available from every network.)

TEOM 1400 - 6 to 12 monthly check of flow and mass determination. Data checked and ratified.

BETA-GAUGE - 6 to 12 monthly check of flow determination and instrument calibration. Data checked and ratified.

Three Stage Impactor - weekly check of flow determination.

Air pollution monitoring in Finland has been decentralized, with many organisations responsible for the measurements in numerous networks. For this reason, QA/QC procedures vary greatly between different networks. At the moment, QA/QC systems for air quality monitoring are under development with work initiated in several networks eg Helsinki Metropolitan Area Network.

Table A2.2: Finland (Total Suspended Particulate)

Site Name	Site Type	Pollutant	Annual Mean μgm^{-3}			98th %ile of daily values μgm^{-3}			99th%ile of daily values μgm^{-3}		
			1992	1993	1994	1992	1993	1994	1992	1993	1994
HARJAVALTA - TORTTILAN KOULU	GI	TSP	26	27			89				
HELSINKI - KAISANIEMI	UT	TSP	57	50	45	175	132	119			
JYVÄSKYLÄ - KESKUSSAIRAA-LANTIE	UT	TSP	49	47		226	218				
JYVÄSKYLÄ - LYSEO	UT	TSP	47	43		219	189				
KOTKA - SUNILA	UB	TSP	66	80			210				
KUOPIO - KESKUSTA	UT	TSP	71	71		244					
KUUSANKOSKI - VALTAKATU	UT	TSP	48	51			179				
LAHTI - TORI	UT	TSP	46	72			303				
LOHJA - LAURINKATU	UT	TSP	51	50	46	246	209	241			
PORI - ITÄTULLI	UT	TSP	81	75			295				
TAMPERE - KESKUSTORI, KIOSKI	UT	TSP	106	113			412				
TURKU - ANINKAISTEN-MÄKI	UT	TSP	257	139		970	417				

QA/QC Information

HIVOL Sampler - measurements made according to National Standard SFS3866: Determination of suspended particulates in the atmosphere. High volume method. (Based on the US Reference Method for the Determination of Suspended Particulates in the Atmosphere (High Volumen Method), National Ambient Air Quality Standards 40 CFR, Part 50, Appendix B).

Three Stage Impactor - weekly check of flow determination.

Table A2.3: France (PM₁₀)

Site Name	Site Type	Pollutant	Annual Mean μgm^{-3}			98th %ile of daily values μgm^{-3}			99th%ile of daily values μgm^{-3}		
			1992	1993	1994	1992	1993	1994	1992	1993	1994
CARLING ECO	UI, UT	PM ₁₀	54	60		100	110				
CLERMONT-F CENT	UT	PM ₁₀	51	53		132	94				
COLOMBELLES STADE	GI	PM ₁₀	42	61		132	181				
FLORANGE HUCK	GI	PM ₁₀	40	51		84	99				
GANDRANGE UNI	GI	PM ₁₀	49	91		84	149				
GIVORS MAIR	UI	PM ₁₀	-	50		-	58				
ILLANGE	GI	PM ₁₀	-	48		-	100				
LA MAX STADE	RURALI ND	PM ₁₀	32	53		58	116				
LYON ETATS	UB	PM ₁₀	55	59		136	90				
LYON PUIITS G	UB	PM ₁₀	60	67		120	119				
MARTIGUES ILE	UI,UT	PM ₁₀	48	52		96	88				
METZ BORN	UB	PM ₁₀	41	50		68	87				
PARIS DANT	UT	PM ₁₀	-	54		-	136				
SAINT-POL/ MER VAN C.	GI	PM ₁₀	55	53		148	153				
THIONVILLE FER B	UI,UT	PM ₁₀	43	78		84	143				

QA/QC InformationAnalyser: β -gauge

Table A2.4: France (Black Smoke)

Site Name	Site Type	Pollutant	Annual Mean μgm^{-3}			98th %ile of daily values μgm^{-3}			99th%ile of daily values μgm^{-3}		
			1992	1993	1994	1992	1993	1994	1992	1993	1994
AURILLAC CEN	UT	BS	39	40		77	87				
BORDEAUX V	UT	BS	30	27		97	76				
GRENOBLE F	UT	BS		64			143				
LA ROCHELLE G	UT	BS	36	37		94	87				
LA ROCHELLE P	UT	BS	50	36			73				
MARSEILLE P	UT	BS	41	38		93	90				
MARSEILLE C	UB	BS	35	40		104	104				
MARSEILLE M	UT	BS	34	24		97	80				
MARSEILLE N	UT	BS	67	77		136	131				
MARSEILLE P	UT	BS	59	62		121	108				
MARSEILLE R	UT	BS	43	45		97	108				
MARSEILLE T	UT	BS	41	41		83	87				
NANTES H	UT	BS		44			111				
PARIS ST J	UT	BS	40	26			75				
ROUEN PTT	UT	BS	52	35		107	93				

QA/QC Information

Table A2.5: Germany

	Site Type	Pollutan t	Annual Mean μgm^{-3}			98th %ile of daily values μgm^{-3}			99th%ile of daily values μgm^{-3}		
			1992	1993	1994	1992	1993	1994	1992	1993	1994
Essen	UR	PM ₁₀		43	42		95	95		109	112
Berlin	UT	PM ₁₀			65			98			123
Hanover	UT	PM ₁₀			36			77			102
Essen - Industrial	UI	PM ₁₀		50	58		128	128		147	136

QA/QC Information

Instrument: β - gauge PM₁₀ at Essen (city + industrial) and Hannover

K10 LVS PM₁₀ Sampler at Berlin

Comparison field tests with PM₁₀ reference sampler (WRAC)

Duplicate measurements with two instruments of same type (to determine precision)

For automatic samplers, QA according to VDI guidelines, 4th General Administrative Instructions (BImSchG) or ISO/DIS 6879

Maintenance and flow control

Comparison field and ring tests

Table A2.6: Ireland

Site Name	Site Type	Pollutant	Annual Mean μgm^{-3}			98th %ile of daily values μgm^{-3}			99th%ile of daily values μgm^{-3}		
			1992	1993*	1994	1992	1993	1994	1992	1993	1994
Dublin	Urban	BS		15			70				
Drogheda	Urban	BS		40			160				
Dundalk	Urban	BS		26			168				
Waterford	Urban	BS		30			114				
Wexford	Urban	BS		39			139				
Limerick C	Urban	BS		38			112				
Limerick Cou	Urban	BS		7			37				
Galway	Urban	BS		21			62				

* winter mean

QA/QC Information

Method : BS1747 OECD Calibration Curve

Table A2.7: Italy

Site Name	Site Type	Pollutant	Annual Mean μgm^{-3}			98th %ile of daily values μgm^{-3}			99th%ile of daily values μgm^{-3}		
			1992	1993	1994	1992	1993	1994	1992	1993	1994
BOLOGNA											
7 sites in 1993		TSP	111 - 153	54 - 137							
FRINZE											
5 sites in 1993		TSP	33 - 45	27 - 77							
GENOA											
24 sites in 1993		TSP	19 - 158	11 - 170							
LIVORNO											
1 site in 1993		TSP	53	44							
MILANO Prov											
15 sites in 1993		TSP	37 - 96	40 - 95							
MILANO City											
3 sites in 1993		TSP	73 - 129	52 - 109							
MODENA											
3 sites in 1993		TSP	107 - 112	106 - 113							
PIACENZA											
3 site in 1993		TSP	77	43 - 70							
RAVENNA											
2 sites in 1993		TSP	63 - 69	76 - 89							

Site Name	Site Type	Pollutant	Annual Mean $\mu\text{g m}^{-3}$			98th %ile of daily values $\mu\text{g m}^{-3}$			99th %ile of daily values $\mu\text{g m}^{-3}$		
			1992	1993	1994	1992	1993	1994	1992	1993	1994
ROMA											
4 sites in 1993		TSP	72 - 126	74 - 101		116 - 123	91 - 164				
SIRACUSA											
3 sites in 1993	indust	TSP	34 - 69	36 - 52							
SASSARI											
1 site in 1993		TSP	22	12							
TRENTO											
6 sites in 1993		TSP	31 - 61	33 - 67							
TORINO											
6 sites in 1993		TSP	123 - 140	104 - 268							
VENEZIA											
5 sites in 1993		TSP	57 - 93	51 - 91							

QA/QC Information

Table A2.8: Luxembourg

Site Name	Site Type	Pollutant	Annual Mean μgm^{-3}			98th %ile of daily values μgm^{-3}			99th%ile of daily values μgm^{-3}		
					1995/6			1995/6			
Luxembourg - city	UT	PM ₁₀			30			61			
Esch/Alzette	UI	PM ₁₀			32			71			

QA/QC Information

Flow controller calibration (software)
 Analogue calibration
 Flow controller calibration (hardware)
 Mass calibration verification

Table A2.9: The Netherlands

Site Name	Site Type	Pollutant	Annual Mean μgm^{-3}			98th %ile of daily values μgm^{-3}			99th%ile of daily values μgm^{-3}		
			1992	1993	1994	1992	1993	1994	1992	1993	1994
Houtakker	PM ₁₀	Regional	49	47	48	110	122	114			
Vredepeel	PM ₁₀	Regional	51	49	44	122	118	118			
Breukelen-S	PM ₁₀	Highway			43						
Eindhoven-G	PM ₁₀	UT		41	43		104	113			
Braakman	PM ₁₀	Regional		44	41	106	137	109			
Den Haag-C	PM ₁₀	UB		41	41	110	126	104			
Rotterdam-C	PM ₁₀	UB			41			97			
Wageningen	PM ₁₀	Regional	47	43	41	133	125	101			
Vlaardingen	PM ₁₀	UT	40	41	40	92	118	106			
Utrecht-C	PM ₁₀	UT		43	40		129	90			
Winjandsr.	PM ₁₀	Regional	40	40	37	89	102	93			
Dordrecht	PM ₁₀	UR			37			100			
Amsterdam	PM ₁₀	UB		41	37		125	92			
Eibergen	PM ₁₀	Regional	41	37	37	96	110	110			
Westmaas	PM ₁₀	Regional	41	41	36	114	129	102			
Wieringerw.	PM ₁₀	Regional	43	32	35	105	98	97			
Apeldoorn-S	PM ₁₀	UT		40	39		118	100			
Witteveen	PM ₁₀	Regional	41	36	33	116	109	116			
De Zilk	PM ₁₀	Regional			31			67			

Regional = outside city boundaries, no nearby sources within 5km

Highway = 25m from busy highway (120000veh/day) outside city boundary

QA/QC Information

Instrument: FAG-FH62-I-N with Anderson 246B PM₁₀ inlet

Accuracy: $\pm 20\%$, Precision: $\pm 10\mu\text{gm}^{-3}$

Intercomparison with gravimetric sampler undertaken

Measurements accredited to EN45001

Table A2.10: Spain (PM₁₀)

Site Name	Site Type	Pollutant	Annual Mean μgm^{-3}			98th %ile of daily values μgm^{-3}			99th%ile of daily values μgm^{-3}		
			1992	1993	1994	1992	1993	1994	1992	1993	1994
Manresa	Urban	PM ₁₀	64	90	89	141	147	177			
Sabadell	Urban	PM ₁₀		88	76		222	140			
Bailen	Urban	PM ₁₀		57	70		116	132			
Jaen	Urban	PM ₁₀		44	49		85	111			
Madrid	Urban	PM ₁₀	39	45	57	120	101	126			
Tarragona	Urban	PM ₁₀	53	57	57	115	130	106			
Sarria de Te (Ge)	Ind.	PM ₁₀	69	59	52	123	124	93			
Los Rosales (hu)	Ind.	PM ₁₀		100	96		183	209			
s. Juan Del p. (hu)	Ind.	PM ₁₀		58	123		200	291			
Bonavista (T)	Ind.	PM ₁₀	82	74	73	189	148	168			

QA/QC Information

Table A2.11: Spain (PM₁₀)

Site Name	Site Type	Pollutant	Annual Mean μgm^{-3} (1/4 - 31/3)				98th %ile of daily values μgm^{-3} (1/4 - 31/3)			
			1992 - 93	1993 - 94	1994 - 95	1995 - 96	1992 - 93	1993 - 94	1994 - 95	1995 - 96
Igualada	-	PM ₁₀	71	58	59	58	108	93	104	95
Martorell	-	PM ₁₀	80	63	57	50	118	101	105	75
Rubi	-	PM ₁₀	146	90	69	63	202	141	118	115
Sabadell	-	PM ₁₀		92	55	74		159	140	97
Sarrià de Ter	-	PM ₁₀	89	72	57	50	96	106	88	78
Tarragona (BB)	-	PM ₁₀	50	43	49	35	77	71	96	61
Tarragona (NU)	-	PM ₁₀	62	53	49	47	92	87	96	78
Tarragona (UL)	-	PM ₁₀		39	36	40		74	53	53
Sant Celoni	-	PM ₁₀	89	97	59		101	160	93	
La Pobla M.	-	PM ₁₀	64	46	48	41	99	86	79	65

Table A2.12: Spain (TSP)

Site Name	Site Type	Pollutant	Annual Mean μgm^{-3} (1/4 - 31/3)				98th %ile of daily values μgm^{-3} (1/4 - 31/3)			
			1992 - 93	1993 - 94	1994 - 95	1995 - 96	1992 - 93	1993 - 94	1994 - 95	1995 - 96
Igualada	-	TSP	99	95	98	92	166	157	155	138
Martorell	-	TSP	133	117	124	108	198	177	208	178
Rubi	-	TSP	147	114	112	91	202	193	179	147
Sabadell	-	TSP	216	165	101	132	462	248	218	189
Sarrià de Ter	-	TSP	105	91	85	75	153	142	122	123
Tarragona (BB)	-	TSP	89	93	110	76	138	158	250	133
Tarragona (NU)	-	TSP	131	138	172	142	219	237	314	229
Tarragona (UL)	-	TSP		75	83	70		149	147	124
Sant Celoni	-	TSP	198	157	118	103	347	285	196	166
La Pobla M.	-	TSP	84	80	83	77	123	123	131	118

Table A2.13: Sweden

Site Name	Site Type	Pollutant	Annual Mean $\mu\text{g m}^{-3}$			98th %ile of daily values $\mu\text{g m}^{-3}$			99th %ile of daily values $\mu\text{g m}^{-3}$		
			1992	1993	1994	1992	1993	1994	1992	1993	1994
Stockholm	UB	PM ₁₀)								
Oxelösund	UB	PM ₁₀) 12-								
Norrköping	UB	PM ₁₀) 16								
Malmö	UB	PM ₁₀)								
Stockholm Hornsgatan	UT	PM ₁₀	35								

QA/QC Information

Instrument - TEOM

Operated according to USEPA practice

Table A2.14: United Kingdom

Site Name	Site Type	Pollutant	Annual Mean μgm^{-3}			98th %ile of daily values μgm^{-3}			99th %ile of daily values μgm^{-3}		
			1992	1993	1994	1992	1993	1994	1992	1993	1994
Cardiff Centre	UC	PM ₁₀	26	31	34	60	65	76	65	70	80
London Bloomsbury	UC	PM ₁₀	30	29	27	68	75	56	75	77	66
Belfast Centre	UC	PM ₁₀	27	32	26	95	79	66	136	86	76
Newcastle Centre	UC	PM ₁₀	28	29	26	67	65	60	70	70	62
Leeds Centre	UC	PM ₁₀		27	26		64	64		70	76
Hull Centre	UC	PM ₁₀			26			56			62
Liverpool Centre	UC	PM ₁₀		29	25		83	68		93	76
London Bexley	S	PM ₁₀			25			52			58
Bristol Centre	UC	PM ₁₀		27	24		64	59		66	65
Birmingham Centre	UC	PM ₁₀	26	26	23	69	66	55	77	71	63
Southampton Centre	UC	PM ₁₀			23			48			52
Birmingham East	UB	PM ₁₀			21			50			60
Leicester Centre	UC	PM ₁₀			21			50			51
Edinburgh Centre	UC	PM ₁₀	23	23	20		43	41		45	42

QA/QC Information

Analyser: R + P TEOM
 QA: 6-monthly check of flow and mass determination
 QC: all data checked and ratified
 Accuracy: N/A
 Precision: $\pm 4\mu\text{gm}^{-3}$

Table A2.15: United Kingdom

Site Name	Site Type	Pollutant	Annual Mean μgm^{-3}			98th %ile of daily values μgm^{-3}			99th %ile of daily values μgm^{-3}		
			1992	1993	1994	1992	1993	1994	1992	1993	1994
Plymouth	UT	BS	13	7		52	20		62	23	
Cardiff	UB	BS	11	10	8	36	31	29	38	39	34
Slough	UB	BS	6	9	9	20	29	24	24	33	32
Stepney, London	UB	BS	8	7	7	21	19	22	24	19	23
Norwich	UB	BS	9	8	8	35	29	27	47	32	30
Stoke-on-Trent	UB	BS	19	15	14	64	43	40	84	46	49
Mansfield Woodhouse	UB	BS	26	21	18	122	67	66	144	74	79
Barnsley	UB	BS	21	17	15	76	53	53	114	66	67
Manchester	UI	BS	20	17	14		64	59		66	65
Newcastle	UT	BS	17	16	14	60	49	38	83	66	42
Glasgow	UB	BS	13	10	8	57	36	29	64	41	42
Belfast	UB	BS	24	19	14	103	61	41	125	87	48
Hull	UB	BS	15	10	10	87	35	49	97	37	60
Liverpool	UB	BS	11	10	8	45	31	34	58	37	39
Leicester	UT	BS	16	11	10	66	33	39	97	36	55

QA/QC Information

The UK currently has 226 sites monitoring black smoke on a daily basis, using the black smoke technique as described in ISO standard 9835: 1993 and British Standard BS 1747 Part 2: 1969. The sample illustrated in the table has been selected to illustrate data for each region of the UK, and where available to allow comparison with PM₁₀ monitoring in the same towns or cities. Summary data is available in 'UK Smoke and Sulphur Dioxide Monitoring Networks - Annual Summary Tables' [Digest of Environmental Statistics, Department of the Environment, HMSO, 1992, 1993, 1994]

8. ANNEX 3: UNCED AGENDA 21 AND AIR QUALITY MONITORING

The 1992 United Nations Conference on Environment and Development (UNCED) has identified environmental degradation in cities as one of the areas which requires immediate attention. Agenda 21, the sustainable development action plan for the 21st century, highlights in several chapters the importance of urban air pollution and calls upon national and local governments and the international community to increase their efforts towards obtaining reliable and accessible data and information on pollutant concentrations, sources and effects. More specifically, Agenda 21 states:

- 1) "Sustainable development requires the availability of accurate and timely information to help decision makers and the general public in making sound decisions";
- 2) "The existing information activities of Earthwatch should be strengthened, especially in the areas of urban air, ... and global atmosphere";
- 3) "International organisations involved in sectoral information collection activities should strengthen and develop guidelines for co-ordinated harmonised national and international data collection and assessment";
- 4) "Appropriate air pollution control technologies should be developed on the basis of risk assessment.....Air pollution control capacities emphasising monitoring networks and enforcement programs should be installed in all major cities".