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Long-range transport of Aerosol Particles

A Literature Review

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EMEP Co-operative Programme for Monitoring and Evaluation of the Long-range Transmission of Air Pollutants in Europe

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Mihalis Lazaridis, Arne Semb and Øystein Hov

Norwegian Institute for Air Research
P.O. Box 100, N-2027 Kjeller, Norway

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1 Introduction

The purpose of the current document is to present and review latest research findings in the area of atmospheric Particulate Matter (PM). In this document we summarize key information on the physico-chemical characteristics of PM, their monitoring, sampling and analyses methodology, their emissions and sources, their concentration in Europe and we provide background information on exposure aspects and related health effects. In addition we provide a summary on the effect of particulate matter on visibility, climate effects, material damage and acidification-eutrophication. The present document is aimed to serve as a scientific draft paper on the EMEP aerosol programme and provide information on the regional component of atmospheric particulate matter.

Long-range transport of gaseous air pollutants has been studied extensively in Europe the last decades under the framework of the European Monitoring and Evaluation Program (EMEP) (EMEP-WHO, 1997; Eliassen and Saltbones, 1983; Tarasson and Tsyro, 1998; Pacyna et al., 1991) and several national and international efforts (Berdowski et al., 1998; EPA, 1996a,b; EU, 1996, 1997; Quality of Urban Air Review Group, 1996; Position Paper on Particles, 1998). Emissions of pollutants rise up in the air due to buoyancy effects, advect downwind, and disperse horizontally and vertical due to turbulence field and prevailing meteorological patterns. The last years there is an extensive research focus on particulate matter (PM) (EPA, 1996; EU, 1996, 1997; EMEP-WHO, 1997, WHO, 1996) mainly because of serious public health risks for susceptible members of the population and risks to sensitive ecosystems. The transport of PM in the atmosphere is similar to that of gaseous pollutants for the fine particle fraction but deviates at larger sizes (coarse particles) due to deposition processes. Therefore, the long-range transport of PM contributes significantly in the background particle mass and number size distribution. However, there is still considerable debate among the scientific community regarding the vertical exchange processes involved and the spatial and temporal scales of atmospheric particle transport.

Airborne particulate matter is a complex mixture of many different chemical species originating from a variety of sources. Composition, morphology, physical and thermodynamic properties of PM varies in different geographical places and have a seasonal variability (Finlayson-Pitts and Pitts, 1986; Seinfeld and Pandis, 1998; EPA, 1996; Position Paper on Particles, 1998; Alpert and Hopke, 1981). Sources of PM can be either primary or secondary in nature. Primary particles can be furthermore divided as anthropogenic or natural depending on their origin. Secondary formed particles in the atmosphere are from both natural and anthropogenic origin and are originating from chemical transformations of gaseous precursors such as sulphur dioxide, nitrogen oxides and VOCs. Recent research studies highlight also the importance of biogenic hydrocarbons (such as terpenes) to the formation of organic aerosols[1] (Hoffman et al., 1997; Kavouras et al., 1998).

In Europe anthropogenic sources are dominant because of the urbanization of many countries and the large number of vehicle and combustion sources (industrial and residential) (Position Paper on Particles, 1998). Natural sources of primary aerosols in Europe include sea spray, fugitive dust (e.g. soil resuspension by the wind), long-range transport of Sahara dust, volcanic and biogenic emissions. There is a consistent pattern of geographical variability in Europe with lower concentrations of PM_{10} in the far north and higher concentrations in the southern countries (Position Paper on Particles, 1998). This is possible due to natural emissions of unsaturated hydrocarbons (including isoprene), which are highly reactive. However, the majority of monitoring data is from urban networks in some parts of Europe and there is no systematic monitoring program with representative rural sites in most countries. In addition, many research studies have been performed in northwestern Europe, where aerosol concentrations between urban and non-urban areas are not exceeded 20% (e.g. Van Der Zee et al., 1998). This can be attributed to high emissions from a dense population area, the small weather variability between the measurement sites and the importance of long-range transport of air pollutants.

Furthermore, in a number of studies on wintertime concentrations of PM_{10} and black smoke in 14 urban and 14 non-urban locations in Europe indicate a relative small difference (on average 22% for PM_{10} and 43% for black smoke) (Hoek et al., 1997). Similar observations on the regional character of particulate matter are reported also in United States (EPA, 1996).

An important characteristic of atmospheric particulate matter is the tremendous variation in size ranging from tens of micrometers to a nanometer size (EPA, 1996; Lin et al., 1993; Covert et al., 1992; Clarke, 1992). For example combustion-generated particles (vehicle emissions, power generation) are ranging in size between 0.003 to 1 µm. Pollens and soil dust is composed of particles mainly above 2 µm, whereas fly ash from coal combustion produces particles ranging from 0.1 to 50 µm. In addition, aerosols in the atmosphere undergo changes in their chemical composition and size. This is due to a variety of physical and chemical processes such as nucleation (new particle formation), condensation, evaporation, coagulation, deposition (both wet and dry), activation due to water and other gaseous species and aqueous phase reactions (Seinfeld and Pandis, 1998; Finlayson-Pitts and Pitts, 1986).

The lifetime of particulate matter in the atmosphere is ranging from a few days to few weeks and these relative long residence times result to small differences between the average total mass of $PM_{2.5}$ between urban and nonurban continental aerosols (Heintzenberg, 1989). Table 1 shows data on mass concentration and composition of tropospheric aerosols as summarized by Heintzenberg (1989). The data show that the average $PM_{2.5}$ mass in remote areas is about 3 times lower than

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[1] Aerosol is defined as a suspension of solid and liquid particles in gas but has used commonly to refer to the particulate component only.

the urban concentration whereas the nonurban continental is 2 times lower. These observations show the importance of the regional component of particulate matter that is attributed to long range transport of pollutants.

Table 1: Mass and chemical composition of tropospheric particulate matter (Heintzenberg, 1989)

Region	Mass ($\mu g\ m^{-3}$)	Percentage Composition					
		Unspecified	C (elem)	C (org)	NH_4^+	NO_3^-	SO_4^-
Remote (11 areas)	4.8	57	0.3	11	7	3	22
Nonurban Continental (14 areas)	15	19	5	24	11	4	37
Urban (19 areas)	32	18	9	31	8	6	28

The chemical complexity of atmospheric aerosols requires a consideration of their composition and sources. In addition, because of the considerable influence of natural sources (with larger size in general than the anthropogenic emissions) (Position Paper, 1998) there is a need for measurements of smaller particles such as $PM_{2.5}$. This standardization will allow a better understanding of anthropogenic influence and further implementation of control strategies.

A detailed presentation of the aerosol size distribution for different environmental conditions is given by Jaenicke (1993) and is summarized in Table 2.

Table 2: Parameters for Model Aerosol Distributions expressed as the sum of three log-normal modes (Jaenicke, 1993).

	Mode I			Mode II			Mode III		
Type	N (/cm3)	Dp (μm)	Log(σ)	N (/cm3)	Dp (μm)	Log(σ)	N (/cm3)	Dp (μm)	Log(σ)
Urban	9.93E4	0.013	0.245	1.11E3	0.014	0.666	3.64E4	0.05	0.337
Marine	133	0.008	0.657	66.6	0.266	0.210	3.1	0.58	0.396
Rural	6650	0.015	0.225	147	0.054	0.557	1990	0.084	0.266
Remote - continental	3200	0.02	0.161	2900	0.116	0.217	0.3	1.8	0.380
Free tropo-sphere	129	0.007	0.645	59.7	0.250	0.253	63.5	0.52	0.425
Polar	21.7	0.138	0.245	0.186	0.75	0.300	3E-4	8.6	0.291
Desert	726	0.002	0.247	114	0.038	0.770	0.178	21.6	0.438

The complexity of the processes controlling the shape and size of airborne particles, the enormous number of different sources and the difficulties of their measurements resulted to critical reactions against proposed national ambient air quality standards in United States (Abelson, 1998). The same critique is expected to arise in European level when future Directives for controlling the PM concentrations in Europe will be proposed. On 31 March 1998, a National Academy of Sciences committee in United States delivered a report that highlights the need for additional research before any new standards on PM are adopted. The report stated, "…the committee concludes that EPA should immediately devote more intramural as well as extramural resources to investigating the relationships between fixed-site outdoor monitoring data and actual human exposures to ambient particulate matter, and to identifying the most biologically important constituents and characteristics of particulate matter through toxicological studies.”

Even though the complexity of the factors affecting exposures to particulate matter are recognized, there is an extensive research effort the recent years in basic aerosol research, in examining their physico-chemical properties and the relationship between exposure to particulate matter and resulting health effects (EPA, 1996). Data from epidemiological studies conducted to date demonstrate associations between ambient particulate concentrations and increased morbidity and mortality, while data from toxicological studies have begun to provide potential biological explanations for these observed associations (EPA, 1996; Pope et al., 1995; Schlesinger, 1995; Schwartz, 1994). Morbidity and mortality and their association with particulate matter exposures occur in fact below the current air quality standards (WHO, 1992; WHO, 1994; WHO, 1996; EPA, 1996b; Position Paper, 1998; The European Auto Oil Programme, 1996; APHEA/PEACE, 1995; Pope et al., 1995; Schlesinger, 1995; Schwartz, 1994; NRDC, 1996; Pope et al., 1992; Schwartz and Dockery, 1992; NTIS, 1997; HMSO, 1995; HEI, 1995).

2 Atmospheric Chemistry and Physics of Particulate Matter

2.1 Introduction

The dynamics of the particulate matter in the atmosphere involve various physical and chemical processes under different time scales. Furthermore, aerosol dynamics involve wide range of particle sizes ranging from few nanometers to several hundred micrometers, different compositions and chemical reactivity and undergo complex physical transformations (nucleation, condensation, coagulation and deposition processes). Aerosols arise from natural and anthropogenic sources and are mixture of primary emissions and secondary species. Crustal material, biogenic matter and sea-salt comprise the majority of natural aerosols. Anthropogenic aerosols are composed of primary emitted soot (elemental carbon) and secondary formed carbonaceous material (organic carbon) and inorganic matter (nitrates, sulphates, ammonium and water). Therefore modelling or measuring atmospheric aerosols involves many challenging tasks and is a fast evolving scientific area (Seigneur et al., 1997, 1999; Lurmann et al., 1997; Jacobson, 1997a,b; Turpin, 1999; Lazaridis and Melas, 1998; Zannetti, 1990; Williams and Loyalka, 1991). Meteorological processes affect substantially the physical and chemical processes of atmospheric aerosols as well as the geographical and temporal variation of their sources (EPA, 1996; Position Paper on Particles, 1998). There is a substantial scientific literature examining various aspects of the physicochemical properties of atmospheric aerosols and an overview is presented in the current chapter.

2.2 Physical and Chemical Processes in the Atmosphere

The determination of the aerosol size distribution is one of the most important aspects involved both in measuring and modelling aerosol dynamics. The diameter of an ambient particle can be determined by various means including light scattering measurements, characterization of the aerodynamic resistance of the particle and measurement of its electrical mobility or settling velocity. There is a necessity to refer to an equivalent diameter independently of the measurement method and therefore the Stokes and the aerodynamic equivalent diameter have been introduced. The aerodynamic diameter is defined as the diameter of a spherical particle with equal settling velocity as the particle under consideration but with material density of 1 g/cm^3 (Hinds, 1982).

However, the size distribution is a time and spatial evolving property of atmospheric aerosols and involves transfer of material through the gas phase, vapour phase and particle phase. Based on a modal classification by Whitby (1978) the aerosol size distribution can be viewed as an addition of several log-normal distributions. These include the Coarse Mode (aerosol mass aerodynamic diameter larger than 3 µm), the Fine Mode (between 1 and 3 µm), the Accumulation Mode (from 0.1 to 1 µm) and the Nuclei Mode (below 0.1 µm). Further distinctions within the various modes are performed lately based on observational data, which show a more detailed structure of the size distribution (Hering and Friedlander, 1982; John et al., 1990; Covert et al., 1990; Wiedensohler et al., 1994). There have been also other classifications of the particle size based on their inhalation characteristics and the particles are classified as inhalable, thoracic and respirable (EPA, 1996).

An important portion of the size distribution is the Ultra-fine part of the Nuclei Mode. The understanding of the physics and chemistry of very small clusters containing few hundred of molecules (Preining, 1998) represents a theoretical and experimental challenge.

The aerosol behaviour in the atmosphere is controlled by internal and external processes. Processes that act within the system boundaries are called internal processes whether processes that at act across system boundaries are called external processes (Whitby, 1990). Internal processes include the coagulation, condensation, evaporation, adsorption/desorption, heterogeneous chemistry and nucleation mechanisms. External processes involve convection, diffusion and the effect of external forces such as thermophoresis (Hinds, 1982).

2.2.1 Physical Processes

2.2.1.1 Nucleation

New particle formation in the atmosphere has been observed in the vicinity of polluted sources (Hegg et al., 1985) and in clean, remote regions (Clarke et al., 1998; Clarke, 1992; Covert et al., 1992). Nucleation bursts (homogeneous nucleation) may be responsible for the occurrence of new particle formation in clean environment where the background aerosol concentration is low (Covert et al., 1992; Lazaridis and Melas, 1998; Clement and Ford, 1999a,b).

Nucleation is the initial stage of a first-order phase transition that takes place in various energetically metastable or unstable systems (Abraham, 1974; Jaecker-Voirol and Mirabel, 1989; Lazaridis and Drossinos, 1997; Clement and Ford, 1999a,b). In the atmosphere, where various condensable vapours exist in low concentrations, binary (two-component) or multicomponent nucleation is the predominant particle-formation mechanism (Kulmala et al., 1995; Lazaridis and Melas, 1998). Even though the homogeneous nucleation process is not an important mechanism for determining the aerosol mass size distribution contributes a large amount of newly formed particles in the atmosphere and shapes the number size distribution.

There is an increasing evidence on the role of small aerosol particles in the human health (Seaton et al., 1995; Schlesinger, 1995, EPA, 1996) and on their effect on climate change (IPCC, 1996), making necessary the better understanding of the dynamics, physics and chemistry of the ultrafine aerosols. Nucleation on the surface of pre-existing aerosol particles (heterogeneous nucleation) is also a favourable process in the atmosphere, since in the formation of critical clusters it is not necessary to have such a high supersaturation as in the homogeneous case (Lazaridis et al., 1991).

Several modelling studies have investigated the new particle formation of sulphate particles under various atmospheric conditions (Kreidenweis and Seinfeld, 1988; Covert et al., 1992; Clarke, 1992; Clarke et al., 1998; Kulmala et al., 1995; Russel et al., 1994; Lazaridis and Melas, 1998; Lazaridis and Skouloudis, 1999; Clement and Ford, 1999a,b). These modelling studies are based on the classical model of homogeneous nucleation (Abraham, 1974) or on semiempirical functions (Binkowski and Shankar, 1995). A serious uncertainty in modelling new particle formation for the sulphuric acid-water system is that there are no consistent available experimental data in the whole range of water and acid activities, and temperature; neither current available models for binary nucleation agree closely with experimental results. One reason for the disagreement between theory and experimental results is the conditions under which the experiments were performed, since the system presents intrinsic experimental difficulties related to its corrosive properties and extremely low vapour pressures. However, the classical theory of binary nucleation follows the experimental trend of the measurements at high relative humidities and these at high acid activities.

The classical theory of nucleation is based on the phenomenological concept of a droplet that is viewed as a group of molecules which interact strongly among themselves and weakly with the rest of the system. According to the classical theory, the nucleating cluster is treated with equilibrium thermodynamics as a macroscopic droplet whose free energy of formation depends crucially on the bulk surface tension. The kinetics by which small clusters of the new phase gain or loose molecules is based on ideas developed in chemical kinetics. It is assumed that clusters grow or shrink via the gain or loss of single molecules, an approximation that is reasonable for condensation at low pressures. However, the classical theory being a phenomenological approach lacks a sound microscopic foundation (Lazaridis and Drossinos, 1997).

The nucleation rate according the classical theory can be expressed as :

$$J = J_o \exp(-\beta F^*),$$

where J_o is a kinetic prefactor, βF^* the free energy of formation of the critical droplet, and $\beta = 1/k_B T$ where k_B is the Boltzmann constant and T is the temperature. The expression for the kinetic prefactor for the binary nucleation may be written in a form similar to the one used in one-component nucleation as follows (Lazaridis and Drossinos, 1997):

$$J_o = \rho_V ABZ,$$

where ρ_V is the total density of condensable vapours, A is the surface area of the droplet, B is the average growth rate, and Z is the Zeldovich non-equilibrium factor.

From the modelling point of view there are available models for binary and unary nucleation in the literature that are already integrated in regional and mesoscale transport models (Binkowski and Shankar, 1995; Lazaridis and Melas, 1998). In addition, various parameterisations have been used for the nucleation rate in atmospheric modelling based on experimental. The problem of new particle formation in the atmosphere is an active field and there are still many unresolved questions to be answered (Clarke et al., 1998; Lazaridis, 1998; Clement and Ford, 1999a,b).

2.2.1.2 Particle Growth

Particle growth is occurring through the mass and energy transfer from/to the vapour to the particle phase. Vapour condensation/evaporation and heterogeneous reactions on the surface or inside the particles are the main mechanisms for particle growth.

The condensation mechanism is a rate-limiting process and the ratio of the mean free path in air to the particle radius (Knudsen number, Kn) is an important factor. When particles are much smaller that the mean free path of the surrounding air (free molecular regime) the transport is controlled by the molecular impingement on the particle surface. When particles are larger than the mean free path (continuum regime) the diffusion is the limiting control mechanism. When the Knudsen number is close to one, the regime is called transition regime. The Boltzmann equation can be used for solving rigorously the condensation problem in the transition regime but since there is not a full solution of the Boltzman equation in the whole regime of Knudsen numbers there have been flux matching approximations such as the Fuchs and Sutugin approach (Fuchs, 1964; Sitarski and Nowakowski, 1979). In the flux matching approximations the noncontinuum effects are limited to a region close to the particle and outside the continuum theory applies (Seinfeld and Pandis, 1998).

One approach to model binary condensation is the use of the Mason equation (Mason, 1971) including transitional correction factors (Lazaridis and Koutrakis, 1997). However, other approaches use an equilibrium method to distribute the mass of inorganic matter between the vapour and particle phase and also inside the particle phase (Pilinis and Seinfeld, 1987). A simplified method which is used in modelling urban and regional aerosols with the Urban Airshed Model (UAM-AERO) adopted the equilibrium method together with a mass allocation method to the size distribution (Lurmann et al., 1997).

The condensation rate of vapour species j onto pre-existing aerosol particles using a modified Mason equation, where the transitional correction factors are included can be expressed as (Lazaridis and Koutrakis, 1997) :

$$\frac{dm}{dt} = \frac{4\pi r(S_\infty - S_{r,j})}{N_M / \beta_M + N_T / \beta_T}$$

with

$$N_M = \frac{R_g T_\infty}{D_\infty M p_s(T_\infty)} \quad \text{and} \quad N_T = \frac{L}{k_B T_\infty}\left(\frac{LM}{R_g T_\infty} - 1\right),$$

where β_M and β_T are the transitional correction factors for the mass and heat transfer, D_∞ is the binary diffusion coefficient, M is the molecular weight of the liquid, L is the latent heat of condensation, S_∞ is the saturation ratio of the gas far from the particle and $S_{r,j}$ is the saturation ratio of gas species j at the particle surface.

In addition to the direct condensation on aerosols there are other important mechanisms responsible for their growth as aqueous phase reactions of activated soluble particles in fogs and clouds (Meng and Seinfeld, 1994; Lurmann et al., 1997; Lazaridis and Skouloudis, 1999). Aqueous phase reaction mechanisms are in agreement with experimental observations of the aerosol size distribution.

Hering and Friendlander (1982) observed two distinct types of sulphate aerosols in Los Angeles with mass median diameters of 0.54 ± 0.07 µm and 0.20 ± 0.02 µm respectively. The first type is consistent with an aqueous phase episode during which sulphuric acid formation took place within the droplets, whereas the second results from homogeneous gas-phase SO_2 oxidation. John et al. (1990) obtained similar results but the frequency of occurrence of the two submicron mass distributions is not well established (McMurry and Wilson, 1983; Hobbs et al., 1985). There have been many field studies examining the effect of clouds and fogs on the aerosol size distribution (Kelly et al., 1989; Altshuller, 1983; Hegg et al., 1993).

A semi-empirical fog model for SO_2 is adopted by the UAM-AERO model (Lurmann et al., 1997) and by Lazaridis and Skouloudis (1999). The fog model is presented in Table 3.

Table 3: Empirical chemical reactions for aqueous phase oxidation of SO_2 (Lurmann et al., 1997).

Chemical reactions	Haze and RH > 70%	Heavy fog
SO_2 (g) + H_2O_2 → H_2SO_4 (g)	k = 0.05 ppm^{-1} min^{-1}	k= 5 ppm^{-1} min^{-1}
SO_2 (g) → H_2SO_2 (g)	k= 0.00033 ppm^{-1} min^{-1}	k= 0.00167 ppm^{-1} min^{-1}

The literature of modelling studies of aerosol growth, condensation kinetics and aqueous phase reactions is vast and is evolving fast the last years. Recent reviews are presented by Pruppacher and Klett (1997), Rogers and Yau (1989), Seinfeld and Pandis (1998), Williams and Loyalka (1991), Fuchs (1964) and Hirschfelder et al. (1954).

2.2.1.3 Coagulation

Aerosols in the atmosphere can collide due to their Brownian motion or due to hydrodynamic, electrical or gravitational forces. This is called coagulation (or agglomeration) mechanism and is very crucial in the development of the size distribution in the atmosphere (Friedlander, 1977). The collision of particles in the atmosphere is given by the Smoluchowski equation that is normally expressed in terms of particle volume coordinates (Williams and Loyalka, 1991). Furthermore, the coagulation equation can be written in a continuous or discrete forms (Williams and Loyalka, 1991; Seinfeld and Pandis, 1998).

Following the discrete representation, as a result of coagulation between particles, particles are both removed from and added to size bins (Lazaridis and Koutrakis, 1997). If two particles of masses m_1 and m_2 coagulate, the mass of the particle formed is $m_3 = m_1+m_2$. If $K_{1,2}\, n_1\, n_2$ is the coagulation rate between particles of masses m_1 and m_2, then : $dn_1/dt = - K_{1,2}\, n_1\, n_2$, $dn_2/dt = - K_{1,2}\, n_1\, n_2$ and $dn_3/dt = K_{1,2}\, n_1\, n_2$. There is a net loss of one particle per coagulation but the total mass is conserved. Generalising the above equations we can obtain :

$$\frac{dn_k}{dt} = \frac{1}{2}\sum_{i+j=k} K(m_i, m_j)\, n_i n_j - n_k \sum_{i=1}^{\infty} K(m_i, m_k)\, n_i \quad ,$$

where i+j=k means that the summation is taken over those grid points for which $m_k = m_i + m_j$.

The theory of particle coagulation is reviewed by Fuchs (1964), Friedlander (1977), Williams and Loyalka (1991) and Seinfeld and Pandis (1998). Recent modelling studies on aerosol dynamics including coagulation includes the works of Megaridis and Dobbins (1990), Gelbart (1990) and Seigneur et al. (1996). The complexity of integrating detailed aerosol dynamics in air quality models resulted to the exclusion of coagulation processes (e.g. Lurmann et al., 1997). However, in regional aerosol modelling studies where the effect of number size distribution is crucial in radiative forcing explicit modelling of coagulation processes is included (Binkowski and Shankar, 1995).

2.2.1.4 Deposition

Aerosols and gaseous species are removed from the atmosphere through the mechanisms of dry and wet deposition. It is a common practise to parametrize the deposition process using the concept of deposition velocity. The deposition velocity is defined as the ratio of the deposition flux of the specified pollutant (Seinfeld and Pandis, 1998) to the pollutant concentration. There are two general approaches to determine the dry deposition velocity. In the first method available experimental data for different aerosol and gaseous species are used. The second method is based on the transfer of material from the atmosphere to the earth’s surface through different resistance mechanisms, the aerodynamic resistance, the surface resistance and the transfer resistance (Slinn and Slinn, 1980).

The particle deposition varies with the particle size and can be expressed as (Slinn and Slinn, 1980):

$$v_d^i = \frac{1}{r_a + r_d^i + r_a r_d^i v_g} + v_g^i$$

where the term at the left hand side is the deposition velocity (m/s) of particles in the size bin i, r with subscript a is the aerodynamic resistance (s/m), r with subscript d is the deposition layer resistance (s/m) of particles in the size bin i, and v with subscript g is the gravitational settling velocity (m/s) of particles in the size bin i.

A review of field measurements for determining the value of deposition velocity of various species is presented by Davidson and Wu (1990). However, the measurements of dry deposition have not yet resolved a full understanding of the dry deposition process since the problem is quite complex and involves many factors that cannot be accounted in the different field studies. These uncertainties include meteorological conditions including temporal and spatial characteristics of atmospheric turbulence, surface characteristics and aerosol properties.

Deposition velocities from various gaseous and aerosols are reported by Sehmel (1980), Nicholson (1988), Pierson et al. (1986), Pierson et al. (1988) and Pierson and Brachaczek (1990).

Wet scavenging is also an efficient mechanism for removal of aerosols from the atmosphere. Below-cloud scavenging has an approximate value of $3x10^{-5}$ s^{-1} with in-cloud scavenging about ten times larger (Hicks and Meyers, 1989; see also for a detailed discussion EPA, 1996). The wet deposition velocity can be expressed as the product of an average scavenging rate (Λ) and the vertical height h (Finalyson-Pitts and Pitts, 1986), where a uniform distribution of pollutant is assumed between the earth surface and height h. A detailed discussion on wet deposition characteristics is presented by Finlayson-Pitts and Pitts (1986). There is an extensive literature on field studies on wet deposition that highlights the importance of atmospheric variability on wet deposition efficiency removal (Slinn and Slinn, 1981; Waldman et al., 1990; Harrison and Pio, 1983; Jaffrezo and Colin, 1988).

2.2.1.5 Modelling Aerosol Dynamics

Two main methods have been elaborated to model the aerosol dynamics using a detailed aerosol-size distribution, namely the sectional and moment methods. The main objective is to solve the General Dynamic Equation (GDE) of aerosols using a comprehensive method to treat the complexity of the aerosol size distribution dynamics (Seinfeld and Pandis, 1998). However, since the inclusion of detailed aerosol dynamic models in mesoscale or regional modelling is a difficult and computational intensive task various simplifications have been made omitting various terms of the GDE (Lurmann et al., 1997; Lazaridis and Melas, 1998).

In the sectional method the size distribution is divided in several size bins (sections) logarithmically spaced. A common assumption is that all the particles in each section have the same chemical composition (internally mixed assumption) (Gelbard., 1990; Seigneur et al., 1986; Jokiniemi et al., 1994). Incorporation of sectional methods into mesoscale and point plume models is a quickly evolved area (Seigneur et al., 1997; Lurmann et al., 1997; Lazaridis and Melas, 1998).

In the moment method the moments of the aerosol size distribution are expressed in terms of the distribution parameters. Predictions of the three moments of the aerosol size distribution results to the determination of the geometric mean diameter and the geometric standard deviation as function of time (Binkowsi and Shankar, 1995; Achermann et al., 1998). The moment method is adopted in the Regional Acid Deposition Model to determine the dynamics of sulphur aerosol over eastern United States (Binkowsi and Shankar, 1995).

2.2.1.6 Wind Erosion

Wind erosion of soil particles due to atmospheric turbulence is the most important pathway for emission of primary particles in the atmosphere (Peterson and Junge, 1971; Gillette and Hansson, 1989). However, the resuspension process is a complex one since it involves particle-surface and turbulence-particle interactions where the morphology of the surface plays a very important role (Reeks et al., 1988; Braaten et al., 1989; Lazaridis et al., 1998; Lazaridis and Drossinos, 1998).

Current models of particle resuspension are based either on fitting of experimental results (Gillette and Hansson, 1989), or on force- or energy-balance models. In force-balance models resuspension occurs when aerodynamic lift forces become greater than the adhesive forces. Reeks et al. (1988) proposed a different approach to particle resuspension by developing a model based on an energy-balance approach. Modifications of the energy-balance model and applications to multilayer resuspension developed by Lazaridis et al. (1998) and Lazaridis and Drossinos (1998).

An overview of field studies on the resuspension problem is presented by Sehmel (1973). Particulate matter emissions due to the resuspension process in global level are estimated to be close to 1,800 to 2,000 x 10^6 tonnes per year. Field studies have shown that dust storms are in general connected with meteorological frontal activity resulting to a long range transport of dust to many 100’s of kilometres (EPA, 1996).

An important aspect of soil resuspension by turbulent flow is connected with the study of radioactive particulate matter transport and dispersion and examining the contribution to any potential risk of human health. The concepts of resuspension rate (J) and resuspension factor (K) are used to describe the process of soil resuspension. The resuspension rate (1/s) can be expressed as (Lazaridis and Georgopoulos, 1998) : $J=Q/c$, where Q is the upward turbulent flux of particulate matter and c is the concentration of particles under study of the underlying soil. An alternative method for measuring the resuspension is based on the concept of the resuspension factor K, which can be expressed as $K = q/c$, where q is the mean atmospheric concentration measured near the surface level. Data for the resuspension factor following the Chernobyl accident were measured in a selection of European sites and a simple model for the resuspension factor $K = A \exp(-B\, t)$ has been fitted to the data, where t is the time in months.

2.2.2 Chemical Processes

2.2.2.1 Gas Phase Reactions

Atmospheric chemistry involves a large number of reactive species which are in ppm and ppb levels. For example the formation of ozone and nitrogen dioxide involves a large number of non-linear chemical reactions (e.g. Simpson, 1995; Gery et al., 1989). Furthermore, the chemical reaction rates depend also on the background concentration of the various chemical species that is determined from the emission and meteorological characteristics. The modelling of gaseous chemical reactions in the atmosphere is a difficult task because of the complex chemical reactions and the stochastic mixing processes due to turbulence. Several simplifications are adopted in describing the gaseous phase chemical reactions in air quality models.

The gas-phase chemical mechanisms included in air quality models do not include an explicit reaction scheme for all chemical reactions but instead use “lumped” categories such as the SAPRC and RADM mechanisms (including lumped mechanisms for alkanes, alkenes etc.) and the CBM-IV mechanism (lumped bonds such as C-C, C=C) (see Table 4).

Table 4: Gas phase chemical mechanisms.

EMEP	Simpson, 1995
CBM-IV	Gery et al., 1988, 1989
RADM2	Stockwell et al., 1990
GATOR	Jacobson, 1997a,b
SAPRC	Carter, 1995, 1996
LCC	Harley et al., 1993

2.2.2.2 Vapour-Particle Equilibrium

The simulation of the vapour/particle equilibrium in the atmosphere involves the prediction of the main primary and secondary components of atmospheric aerosols in the whole range of the particle size distribution. The main components of aerosols include sulphate, nitrate, ammonium, chloride, elemental carbon, organic carbon, water, chloride and crustal material (Lurmann et al., 1997). Incorporation of a simplified treatment of aerosol chemistry in combination with existing air quality models has been already performed (Seigneur et al., 1997). Various parameterizations have been adopted in modelling secondary organic matter formation and inorganic aerosol concentrations and an overview is presented by Seigneur et al. (1997).

The formation of sulphuric acid in the gaseous phase is mainly concentrated on the oxidation of sulphur dioxide by hydroxyl radicals :

$$SO_2 + OH \rightarrow H_2SO_4 + HO_2$$

The produced sulphuric acid mainly condenses on pre-existing aerosol particles or leads to new particle formation due to homogeneous nucleation. The production of secondary organic particulate matter and of gaseous low-volatility organic products is generally described using a condensed lumped reactions including an oxidant reactant (e.g. OH, O3, NO3) (Seinfeld and Pandis, 1998) :

$$HC + Oxidant \rightarrow xX + yY + ... + gG$$

where X, Y are the gaseous phase organic products and G refers to condensable organic gas that forms secondary organic aerosols. The low case characters (x,y,…,g) are the corresponding stoichiometric coefficients that correspond to aerosol yields.

One of the widely available models for inorganic aerosols is the SEQUILIB equilibrium model that predicts the gas-phase and aerosol-phase concentrations of various species (Pilinis and Seinfeld, 1987). The SEQUILIB model includes the following species:

$$NH_4Cl, NH_4NO_3, (NH_4)_2SO_4, NH_4HSO_4, (NH_4)_3H(SO_4)_2$$

$$NH_3, HCL, HNO_3, H_2O, NH_4^+, SO_4^=, NO_3^-, Na^+, Cl^-, HSO_4^-, H_2SO_4,$$
$$Na_2SO_4, NaHSO_4, NaCl, NaNO_3$$

The concept of thermodynamic equilibrium is widely used in modelling gaseous and particulate phase reactions of inorganic and organic nature. Alterations of hygroscopic properties of aerosols due to incorporation of secondary organic matter in the particle phase is not incorporated in currently in use air quality models (Saxena et al., 1995).

2.3 Chemical Composition

2.3.1 Acid Aerosols-(Sulphates-Nitrates)

Aerosol acidity is defined as acids and their precursors residing in the atmosphere in the gas and aqueous phase (Sioutas and Koutrakis, 1996). Strong acidity refers mainly to sulphuric acid or partly neutralized acid particles, whereas weak acidity includes a number of inorganic (e.g. nitrous acid, hydrogen phosphates, hydrochloric acids) and organic species (e.g. phenols, carboxylic acids). Even though in the atmosphere weak acids are not predominant, may occur indoors (Lawrence and Koutrakis, 1994; Waldman et al., 1990; Koutrakis et al., 1989; Zhang et al., 1994). Field studies indicate a spatial homogeneity in particle strong acidity over large geographical areas and is occurring mainly during warmer periods and many times occurs at the same time as photochemical smog episodes (Sprengler et al., 1989; Thurston et al., 1992).

The production of sulphuric acid in the atmosphere occurs mainly through photo-oxidation of sulphur dioxide with hydroxyl radicals in the gaseous phase. Aqueous phase reactions of SO_2 with hydrogen peroxide (H_2O_2) is also a very important pathway for sulphuric acid production since it occurs also at nighttime. Further neutralization of the aerosol particles occur with the diffusion of ammonia (NH_3) in the liquid phase (Sioutas and Koutrakis, 1996).

Sulphate formation in clouds and fogs, where aqueous phase reactions is dominant process has studied by several researchers (e.g. Seigneur and Saxena, 1984; Joos and Baltensprenger, 1991; Pandis et al., 1990). High production rates of aerosol particles is observed in the vicinity of clouds suggesting homogeneous nucleation of H_2SO_4/H_2O particles (Hegg et al., 1993; Clarke et al., 1998; Clement and Ford, 1999a,b).

Nitric acid is mainly formed through the photooxidation reaction between NO_2 and OH. During night time there is a reaction between NO_2 and O_3 to form NO_3 which is further reacts with NO_2 to form nitrogen peroxide (N_2O_5). Furthermore, N_2O_5 can react with water vapour to form aqueous nitric acid. Even though this is a slow reaction it can be reaction of N_2O_5 with condensed water (cloud or fog droplets) that is considerably faster (Tuazon et al., 1983). Nitric acid can be neutralized when it is reacting with ammonia and forms particulate ammonium nitrate. Nitric acid can be also react with salts of chlorine or carbonate and forms particulate salt solution. In addition the reactions in the atmosphere between NO and OH leads to the production of nitrous acid (HONO).

Calculations for the inorganic portion of the atmospheric aerosols is mainly performed using multicomponent equilibrium models. An example of this kind is the SEQUILIB equilibrium model that calculates the total quantities of ammonium, chloride, nitrate and water components of the atmospheric aerosols

(Pilinis and Seinfeld, 1987; Pandis, 1996). The SEQUILIB model is also integrated in the mesoscale air quality model UAM-AERO (Lurmann et al., 1997).

2.3.2 Carbon-Containing Aerosols

2.3.2.1 Elemental Carbon- Primary Organic Carbon

Elemental carbon (EC) has a chemical structure similar to impure graphite and is emitted as primary particles mainly during combustion processes (wood-burning, diesel engines (EPA, 1996; Burtscher, 1992; Hansen and Lacis, 1990). In Western Europe the contribution of diesel emissions to EC concentrations is estimated to be between 70 and 90% (Hamilton and Mansfield, 1991). Elemental carbon both absorbs and scatters light and contributes significantly to the total light extinction (Finlayson-Pitts and Pitts, 1986). Much higher concentrations of EC are found in urban areas compared to rural and remote locations. In rural and remote locations the EC concentration vary between 0.2 to 2.0 $\mu g\ m^{-3}$ (Clarke et al., 1984; Pinnick et al., 1993) and between 1.5 to 20 $\mu g\ m^{-3}$ in urban areas (Heintzenberg and Winkler, 1984; Rau, 1989). Clarke (1989) found that the concentration of EC in remote ocean areas is ranging between 5 to 20 ng m^{-3}. Studies concerning the size distribution and characteristics of EC aerosols is summarized by Burtscher et al. (1993), Venkataraman and Friedlander (1994) and Chow et al. (1994). Average EC concentrations are around 1.3 and 3.8 $\mu g\ m^{-3}$ for rural and urban sites respectively in U.S. The ratio of EC to total carbon is ranging between 0.15 to 0.20 in rural areas and between 0.2 to 0.6 in urban areas (Wolff et al., 1982; Chow et al., 1993).

The organic carbon is a complex mixture of thousands of different organic compounds (Cass et al., 1982; Turpin and Huntzicker, 1995; Grosjean, 1992; Odum et al., 1997; Pandis et al., 1992) and a very small portion of it is molecular characterized (around 10%). Organic compounds that have been characterized include among others n-alkanes, n-alkanoic acids and polycyclic aromatic compounds. Due to difficulty in measuring organic compounds our current knowledge about organic matter is limited and incomplete. Primary emission sources for organic carbon include combustion processes, geological (fossil fuels) and biogenic sources.

2.3.2.2 Secondary Organic Matter Formation (Secondary Organic Carbon)

An important part of secondary aerosol particles in the atmosphere is composed by secondary formed organic matter (Turpin and Huntzicker, 1991) produced from oxidation of organic compounds. Partitioning of gas-particle organic compounds in the atmosphere is an important task for determining their association with the fine particulate matter. Understanding the mechanisms that control the conversion of organic matter from the vapour to particulate matter will provide valuable information for determining future control strategies for reducing the partition of organic matter in the particulate phase. However, there is a great complexity of the number of different chemical forms of organic matter and absence of direct chemical analysis which resulted to use mainly experimentally determined fractional aerosol yields, fractional aerosol coefficients and adsorption/absorption methodologies (Cass et al., 1982; Turpin and Huntzicker, 1995; Grosjean, 1992; Odum et al., 1996, 1997; Pandis et al., 1992;

Pankow, 1987; Lazaridis, 1999) to describe the incorporation of organic matter in the aerosol phase.

An important pathway for secondary organic particle formation is arising from biogenic hydrocarbons. There are very large quantities of biogenic hydrocarbons that are globally emitted which are also highly reactive (Hoffmann et al., 1997). Annual global emissions of biogenic hydrocarbons are estimated to be between 825 and 1150 Tg C (per year), whereas the anthropogenic emissions are estimated to be less than 100 Tg C (per year). A detailed overview of the formation of organic aerosols from biogenic hydrocarbons is reviewed by Hoffmann et al. (1997).

An empirical approach to describe secondary organic aerosol (SOA) formation is based on the fractional aerosol coefficient (FAC) method that is defined as (Grosjean and Seinfeld, 1989):

$$\text{FAC (dimensionless)} = [\text{SOA}]\ (\text{mg m}^{-3})/[\text{VOC}]_o\ (\text{mg m}^{-3})$$

where $[\text{VOC}]_o$ is the initial VOC concentration. The VOC concentration is expected to be obtained by experimental data and the SOA mass concentration from available smog chamber experiments (based on VOC-NO_x irradiation and VOC-O_3 reaction in dark). Therefore, the FAC is defined through a number of empirical parameters which is not taking into account SOA variations based for example on VOC/NO_x ratio. Application of FAC for individual VOC is estimated by Grosjean (1992).

Aerosol yield is defined as the fraction of a reactive organic gas that is transformed to aerosol. There have been several methods for describing the partition of organic matter into the aerosol phase. Pandis et al. (1992) placed the condensable organic material to the aerosol phase when their gas-phase concentration exceed their equilibrium vapour pressure. Assuming a negligibly small saturation vapour pressure resulted to condense the majority of the condensable organic matter in the particulate phase. Pankow (1994) suggested that, even undersaturated organic gaseous matter can participate in the aerosol phase using the mechanisms of adsorption and absorption processes (Pankow, 1994) for determining the gas-particle partitioning of organic matter in the atmosphere. Applications of the adsorption/absorption and aerosol yields mechanisms in atmospheric conditions are performed by Lazaridis (1999).

A comprehensive chemical mechanism where the condensable organic compounds (COC) yields for the lumped organic compounds are obtained by Pandis et al. (1992) is incorporated in the Carbon Bond IV (CB-IV) chemical mechanism (Gery et al., 1988). A number of 16 reactions which are included in the CB-IV mechanism are presented in Table 5. The list of species involved in the chemical reactions is presented in Table 6.

Table 5: A lumped chemical scheme for COC yields.

Reactants	Products (COC refers to (μg m^{-3})	Rate constant (ppm^{-n} min^{-1})
O + OLE2	0.63ALD2+0.38HO2+0.28XO2 +0.3CO+0.2FORM+0.02XO2N +0.22PAR+0.2OH+1236COC	5.92E3
OH+OLE2	FORM+ALD2+XO2+HO2-PAR+1236COC	4.2E4
O3+OLE2	0.5ALD2+0.74FORM+0.22XO2 +0.1OH-PAR+1236COC	1.8E-2
NO3+OLE2	0.91XO2+0.09XO2N+FORM +ALD2-PAR+NO2+1236COC	1.14E1
MEOH+OH	FORM+HO2	1.6E3
ETOH+OH	ALD2+HO2	4.3E3
PAR+OH	0.87XO2+0.13XO2N+0.11HO2 +0.11ALD2+0.76ROR-0.11PAR +8COC	1.2E3
O+OLE	0.63ALD2+0.38HO2+0.28XO2 +0.3CO+0.2FORM+0.02XO2N +0.22PAR+0.2 OH + 20COC	5.92E3
OH+OLE	FORM+ALD2+XO2+HO2-PAR +20 COC	4.2E4
O3+OLE	0.5ALD2+0.74FORM+0.33CO +0.44HO2+0.22XO2+0.1OH -PAR+20COC	1.8E-2
NO3+OLE	0.91XO2+0.09XO2N+FORM +ALD2-PAR+NO2+20COC	1.14E1
OH+TOL	0.08XO2+0.36CRES+0.44HO2 +0.56TO2+402COC	9.15E3
OH+CRES	0.4CRO+0.6XO2+0.6HO2 +0.3PEN+221COC	6.1E4
NO3+CRES	CRO+HNO3+221COC	3.25E4
OH+XYL	0.7HO2+0.5XO2+0.2CRES +0.8MGLY+1.1PAR+0.3TO2 +416COC	3.62E4
MTBE+OH	1.37XO2+0.98HO2+0.42FORM +0.97PAR+0.02XO2N	4.18E3

However, there are several limitations and uncertainties connected with the SOA modelling and understanding of the organic matter formation in the atmosphere. Seigneur et al. (1997) summarized the existing uncertainties into five categories:

- Uncertainties that are inherent to the definition and use of the FAC. This is mainly due to the fact that FAC is a single, constant parameter assigned to each VOC.
- Uncertainties in the gas-phase chemistry mechanisms used in air quality models. Some of these uncertainties have a direct impact on the description of the SOA formation.
- Weaknesses in the treatment of SOA in air quality models. For example an important uncertainty is arising due that SOA yield approach is limited to the OH-VOC reaction with ignoring the ozone-VOC reaction.
- Gaps in emission data for those VOC that are SOA precursors.
- Lack of experimental data for evaluation of model performance.

Table 6: List of species used in the lumped chemical scheme for COC yields.

OLE2	Monoterpenes with Olefinic Bond
ALD2	Aldehyde
MEOH	Methanol
ETOH	Ethanol
MTBE	Methyl Tertiary Butyl Ether
XO2	Peroxy radical for NO to NO2 conversion
OH	Hydroxyl Radical
HO2	Hydroperoxy Radical
FORM	Formaldehyde
CO	Carbon Monoxide
XO2N	Peroxy radical for NO to RNO3 conversion
ROR	Lumped Peroxy Radicals
COC	Condensable Organics
O	Ground State Oxygen atom
OLE	Olefinic Bond
MGLY	Methyl Glyoxyl
CRES	Cresols
PAR	Paraffinic Bond

2.3.2.3 Metals and Other Trace Elements

Trace metals in atmospheric particulate matter are mainly from anthropogenic sources such as residential wood combustion, forest fires, combustion of coal and oil, waste incineration and metal smelting (Chow et al., 1992). In the fine particle size range there have been found various trace metals including mainly Pb, Zn, Cd, As, Sb, Ag, In, La, Mo, I, and Sm (EPA, 1996). In the coarse mode there have been found mainly Ca, Al, Ti, Mg, Sc, La, Lu, Hf and Th (Klee, 1984; Bernstein and Rahn, 1979). Furthermore, in both the fine and coarse modes there have been found Na, K, Fe, V, Cr, Co, Ni, Mn, Cu, Se, Ba, Cl, Ga, Cs, Eu, W, and Au (EPA, 1996). Emissions, meteorology and photochemistry are important aspects which control the ambient concentration of trace species in the particulate phase (Finlayson-Pitts and Pitts, 1986). The concentration of trace species found in remote, rural and urban sites are summarised in Table 7.

The importance of long-range transport on the ambient concentration of trace metals is very significant as reported by Schroeder et al. (1987) and Pacyna (1996). Long-term measurements of pollution in North Sea and Baltic Sea indicate that as much as 50% of lead and mercury and 30-50% of arsenic, cadmium, chromium, copper, nickel, and zinc in the water is due to the air deposition process (Pacyna, 1996).

Recent literature concerning with trace metal concentrations in ambient aerosols is given by Schroeder et al. (1987), Pacyna (1996), Klee (1984), Bernstein and Rahn (1979), Morandi et al. (1991), Watson et al. (1994), Chow et al. (1993), Finlayson-Pitts and Pitts (1986) and Harrison et al. (1981).

Table 7: Concentrations (ng m^{-3}) and size distribution of various elements found in atmospheric particles (Schroeder et al., 1987).

Elements	Remote	Rural	Urban (USA)
As	0.007 – 1.9	1.0 – 28.0	2.0 – 2,320
Cd	0.003 – 1.1	0.4 – 1,000	0.2 – 7,000
Ni	0.01 – 60.0	0.6 – 78.0	1.0 – 328
Pb	0.007 – 64.0	2.0 – 1,700	30 – 96,270
V	0.001 – 14.0	2.7 – 97.0	0.4 – 1,460
Zn	0.03 – 460.0	11.0 – 403	15 – 8,328
Co	0.001 – 0.9	0.08 – 10.1	0.2 – 83
Cr	0.005 – 11.2	1.1 – 44.0	2.2 – 124
Cu	0.029 – 12.0	3.0 – 280	3 – 5,140
Fe	0.62 – 4,160	55 – 14,530	130 – 13,800
Hg	0.005 – 1.3	0.05 – 160	0.58 – 458
Mn	0.01 – 16.7	3.7 – 99	4.0 – 488
Se	0.0056 – 0.19	0.01 – 3.0	0.2 – 30
Sb	0.0008 – 1.19	0.6 – 7.0	0.5 - 171

2.3.3 Biological Aerosols

A definition of the primary biological aerosol particles (PBAP) can be written as: “Primary Biological Aerosol Particles describe airborne solid particles (dead or alive) that are or were derived from living organisms, including microorganisms and fragments of all varieties of living things”. PBAP include viruses ($0.005\ \mu m < r < 0.25\ \mu m$) (r, refers to particle radius), bacteria ($r \geq 0.2\ \mu m$), algae, spores of lichen mosses, ferns and fungi ($r \geq 0.5\ \mu m$), pollen ($r \geq 5\ \mu m$), plant debris like leaf litter, part of insects, human and animal epithelial cells (usually $r > 1\ \mu m$) (Matthias-Maser, 1998).

The composition of the PBAP is changing through the year as following:

- Spring : microorganism, pollen, some spores, a few fragments,
- Summer : microorganism, pollen, spores, a few fragments,
- Autumn : microorganism, fragments, spores, a few pollen,
- Winter : microorganism, fragments, spores, some pollen.

2.4 Status of Modelling Aerosol Processes

There are currently several three-dimensional Eulerian air quality models that include a PM module. The modelling of particulate matter is mostly concentrated in a revised chemistry module and deposition module. There is no available aerosol dynamics modelling, no cloud chemistry and no subgrid treatment of subgrid modelling. However, the RPM model has the ability to model detailed aerosol dynamic processes using a modal method to describe the aerosol size distribution (Binkowski and Shankar, 1995).

The description of the pollutant transport in the 3-D air quality models is performed using the atmospheric diffusion equation including a source term and a scavenging term. The atmospheric diffusion equation can be written as (Seinfeld and Pandis, 1998) :

$$\frac{\partial c_i}{\partial t} + u_j \frac{\partial c_i}{\partial x_j} = \frac{\partial}{\partial x_j}\left(K_{jj} \frac{\partial c_i}{\partial x_j} \right) + R_i(x,t) + S_i(x,t)$$

where c_i is the mean concentration of species i, x_j the space coordinate at the direction j (j=1-3), K_{jj} is the eddy diffusivity at the direction j, $R_i(x,t)$ denotes the scavenging of species during chemical reactions and deposition, and the $S_i(x,t)$ is the source term (emissions). The numerical evaluation of the atmospheric diffusion equation is usually performed using an operator splitting technique, which employs a separation of the horizontal and vertical transport terms from the chemical reaction and emission terms so that each term is solved separately.

The available 3-D air quality models including modelling of aerosol processes (in addition to EMEP work) are presented in the next subsections (Seigneur et al., 1997, 1999).

2.4.1 Mesoscale Models

(1) The California Institute of Technology model (CIT)
(2) The Denver Air Quality Model (DAQM)
(3) The Gas, Aerosol, Transport and Radiation model (GATOR)
(4) The Regional Particulate Model (RPM)
(5) The SARMAP Air Quality Model with aerosols (SAQM-AERO)
(6) The Urban Airshed Model Version IV with aerosols (UAM-AERO)

2.4.2 Long Range Transport Models

(7) The Regulatory Modelling System for Aerosols and Deposition (REMSAD)
(8) The Urban Airshed Model Version IV with linearized chemistry (UAM-LC)
(9) The Visibility and Haze in the Western Atmosphere model (VISHWA)

In the European level the EUTREND model (Eerens et al., 1996) has been also used for calculating primary PM_{10}. However, the model has not the possibility to include the formation of secondary aerosols.

Currently, the EMEP model is the main regional regulatory modelling tool in studying photochemical pollutants and acidification Europe and treats the PM_{10} and $PM_{2.5}$ fractions of the atmospheric sulphate and nitrate aerosol using a simplified approach (Tarrason and Tsyro, 1998; Tarrason et al., 1998). The EMEP Lagrangian model calculates the air concentrations of sulphate, nitrate, ammonium sulphate and ammonium nitrate secondary formed particles using lumped reaction rates.

2.5 Effects of PM on Ecosystems, Climate and Materials

2.5.1 Acidification-Eutrophication

Pollutants emitted to the atmosphere such as SO_2 and NO_x are oxidized to sulphate and nitrate through gaseous and aqueous phase reactions. These particle species are removed by both dry and wet deposition to the earth's surface leading to effects such as acidification and eutrophication (mostly due to fertilizers and particle deposition). The deposition of sulphate and nitrate particles is dependent on their size that is controlled by the aerosol dynamic processes in the atmosphere. Eutrophication is becoming a serious threat to coastal environments and seems to be a global problem in the next decades. Water enriched with nutrients leads to higher production of organic matter and results to oxygen deficiency which kills marine life (EMEP-WMO, 1997; Spengler et al., 1989; Pelley, 1998).

2.5.2 Visibility Reduction

Visibility degradation is one of the most readily perceived impact of fine particulate matter. Fine particles absorb and scatter the light and therefore reducing visibility. The process can be described with the Mie theory (Seinfeld and Pandis, 1998). For example in many parts of the United States the visual range has been reduced 70% from natural conditions. In the eastern part of US, the current range is 14-24 miles vs. a natural visibility of 90 miles. In the western US the current range is 33-90 miles vs. a natural visibility of 140 miles. Fine particles (mainly in the accumulation mode, with diameter between 0.3-1.0 μm) make the major contribution to visibility reduction. Recent reviews concerning visibility reduction include the U.S. EPA (1996) report, the National Acid Precipitation Program (Trijonis et al., 1991) and the National Research Council (1993) reports. Furthermore, there is extensive peer-review literature concerning with visibility and aerosols (van de Hulst, 1981; Gabruk et al., 1999; Seigneur et al., 1997; Atkinson and Lloyd, 1984; Richards et al., 1985; Appel et al., 1985; Eldering et al., 1993; Giorgi and Visconti, 1989; Hegg et al., 1993; Hobbs and Radke, 1992; Kerker and Aden, 1991; White et al., 1994).

2.5.3 Radiative Forcing

Particulate matter influences the climate directly (through scattering and absorption of the solar radiation) and indirectly through the formation of cloud condensation nuclei. The direct aerosol contribution to radiative forcing is due to sulphate aerosols, fossil fuel soot and biomass burning. The radiative forcing due to sulphate aerosols is estimated to be -0.4 W m^{-2}, with a factor of two uncertainty (IPCC, 1996). The effect of soot aerosols is + 0.1 W m^{-2} with a factor of three uncertainty and the contribution from biomass burning is estimated to be -0.2 W m^{-2} with a factor of three uncertainty. Therefore the total direct forcing is estimated to be -0.5 W m^{-2} with a factor of three uncertainty. The indirect effect of aerosols is still uncertain with an estimated effect from 0 to -1.5 W m^{-2} (IPCC, 1995). In addition, aerosols can result to considerable changes of soil moisture which can have impacts in the hydrological cycle on vegetation (IPCC, 1995). Recent peer-reviewed literature concerning radiative forcing and atmospheric aerosols include the works of Charlson et al. (1987, 1991, 1992), Albrecht (1989), Coakley et al. (1983), Davies (1993), Hansen and Lacis (1990), Hoffman et al.

(1994), Penner et al. (1993, 1994), Robock (1991), Myhre et al. (1998), and Schwartz (1988).

2.5.4 Soiling and damage to Materials

An important effect of particulate matter pollution is the soiling of man-made surfaces.

The process of cleaning, painting and repairing exposed surfaces becomes an economic burden. Acid particles can severely deteriorate art works and historic monuments (cultural heritage) and results to the reduction of their aesthetic appeal and life span. Chemical degradation of materials due to deposition of atmospheric acid particles is an important aspect of material damage. Extensive literature concerning material damage due to atmospheric aerosol pollution can be found in the works of Baedecker et al. (1992), Butlin et al. (1992), Cobourn et al. (1993), Hamilton and Mansfield (1993), Haynie and Lemmons (1990), Ligocki et al. (1993), Nazaroff and Cass (1991), van Aalst et al. (1986), Williams (1988).

3 Recommendations

3.1 Modelling

Much of the needed research for sulphur, tropospheric ozone and fine particulate matter overlaps. Atmospheric oxidation reactions are important also in particle formation besides to ozone. Modelling the transport and fate of sulphur, ozone and particles relies on similar meteorological processes, the same computational framework, similar emission inventories and model initializations.

However, there is a number of necessary steps required for performing a realistic modelling of particulate matter. The following points have to be addressed for future modelling studies of PM :

(1) Emission inventories for primary particulate matter (size/chemical composition resolved). A valuable study concerning the particulate matter emission inventory during 1990 and 1993 for PM_{10}, $PM_{2.5}$ and $PM_{0.1}$ is published recently (Berdowski et al., 1996).

(2) Modifications in the Gaseous Chemistry modules including a new treatment of secondary organic particulate formation, sulphate and nitrate formation in the presence of fog and/or clouds.

(3) Incorporation of aerosol dynamics modules such as nucleation, condensation, size resolved deposition and inorganic aerosol equilibrium model. Aerosol dynamics is a necessary step for obtaining a realistic size distribution/chemical composition of the particulate matter in the atmosphere. Size/composition are important factors for determining the particle radiative forcing of aerosols, health effect consequences and composition of the deposited particles.

(4) Characterization of the aerosol size distribution/chemical composition in Europe. This information is necessary for using it as background and initial conditions in future simulations.

(5) Use a modified version of a mesoscale model such as UAM-AERO for detailed modelling of particulate matter dynamics in a particular area of Europe (e.g. South Norway/Denmark/South Sweden). This task could require meteorological inputs in a fine grid scale (5 x 5 km^2). Using a mesoscale aerosol model as a regulatory tool we can determine the air concentration/size distribution/chemical composition/deposition profile of particulate matter in different parts of Europe.

The UAM-AERO is an air quality model which simulates the atmospheric processes governing ambient concentrations of PM (Lurmann et al., 1997). The model simulates emissions of particle and gaseous species into the atmosphere, horizontal and vertical transport of pollutants, dry deposition and chemical reactions. The UAM-AERO is an extension of the well known photochemical model UAM-IV. It uses an extended version of the CB-IV chemical mechanism (or the SAPRC90 as an alternative) where the chemistry of secondary formed aerosols is treated. The model predicts the following chemical components of PM:

nitrate, sulphate, ammonium, sodium, chloride, elemental carbon, organic carbon, crustal material and water.

(6) Treatment of subgrid phenomena. An important problem in the modelling of urban and regional photochemical pollution systems is related to the discretized representation of the physical airshed by the numerical modelling grid. For example aerosol yields obtained from experimental measurements and estimates also indicate that there are highly non-linear aspects involved in the production of organic aerosols (e.g. the yields for the monoterpenes and peroxyl radicals). Therefore, the subgrid scale variability of several organic aerosol species is expected to have very significant effects on the modelling estimates of air quality.

3.2 Monitoring/Measurements

(1) Use of the pre-existing Ozone Monitoring Stations network for placing $PM_{2.5}$ and PM_{10} monitors. This will reduce cost, will reduce burdens, and achieve multiple monitoring objectives. However, the PM measurement network need to be maintained at a daily resolution for determining actual exposure of human receptors.

(2) Place additional monitoring stations in areas with poor air quality and high population density (urban areas, industrial zones).

(3) Determine a reference method for $PM_{2.5}$.

(4) Design a monitoring network with core community-oriented monitors (continuously sampling or everyday sampling) and supplementary monitors that will be allowed to sample less frequently.

(5) Chemical analysis for the core-monitoring samples.

In addition to the above recommendations the EMEP Working Group on “Assessment of effects” (EMEP-WMO, 1997) has highlighted the following measurement needs:

(6) Measurements of base cations in aerosols need to be added to the existing air chemistry measurements programmes.

(7) An effort is needed for separating gaseous and aerosol nitrogen species since their relative behaviour and contribution to effects differ.

4 Monitoring, Sampling and Analysis of Particulate Matter

4.1 Introduction

There are several requirements for realistic sampling and analysis of atmospheric aerosols. Special consideration must be given to the following characteristics:

- Air intake efficiencies.
- Size-selective sampling.
- Sampling artefacts, changes of gas-particle partitioning.
- Sampling substrate interferences with the chemical analysis method.
- Determination of mass by weighing or by other methods.

In addition, considerations must be given to the available analysis methods, the representativity of the sampling site in relation to human exposure or the general concentration level of the area or region, depending on the purpose of the investigations.

Overviews of the various measuring techniques used in air quality sampling can be found in the works of Bruynseels et al., 1988, Biswas et al. (1987), Buckley et al. (1991), Cadle et al. (1983), Edwards et al. (1983), Daisey et al. (1982), Eatough et al. (1993), Federal Register (1978), Holländer et al. (1988, 1990), Horvath and Kaller (1994), John and Wang (1991), Köning et al. (1980), Larssen and Hagen (1996), Larssen and Lazaridis (1998), Mark et al. (1992), Marple (1978), McFarland et al. (1977), Nevalainen et al. (1992), Pankow (1987), Rodes et al. (1985), Watson et al. (1989) and Whitby (1978).

The following sections will briefly discuss the principles of aerosol sampling and some of the most important artefacts of common sampling methods.

4.1.1 Construction of air intakes and air intake efficiencies

Most aerosol sampling methods involve drawing the aerosols through sampling ducts or tubes. In outdoor air, isokinetic sampling is generally not possible and non-isokinetic sampling may result in low collection efficiency for particles which have aerodynamically equivalent diameters of more than a few μm. The collection efficiency of non-isokinetic sampling heads in different configurations is a classical problem in aerosol fluid mechanics(e.g. Davies, 1968; Agarwal and Liu, 1980). Sampling efficiencies of inverted-funnel air intake arrangements in relation to isokinetic sampling heads have been investigated in wind tunnels (Steen and Andreasson, 1974; Steen and Johansson, 1975; Barrett, Ralph and Upton, 1982). These studies show that many air intakes will have seriously reduced sampling efficiencies for particles larger than 5-10 μm a.e.d., and that this sampling inefficiency increases with increasing wind speed. Improvement in sampling head design are possible by replacing the inverted funnel intakes by aerodynamically more satisfactory designs. However, as shown by Wedding et al. (1977) and others, representative sampling of particles larger than 10-15μm a.e.d. is very difficult to achieve.

Restricting the sampling to particles less than 10 μm a.e.d. by incorporating an impactor stage or a cyclone in the sampling inlet will reduce these problems. The sampling inlet initially developed with a 15 μm a.e.d. impactor by Liu and Pui at the University of Minnesota and Loo at the Lawrence Berkeley laboratory (Shaw et al., 1983) has been extensively applied after modification to a cut-off at 10 μm.

The European Community (EU, 1999) has decided to adopt two reference samplers for the determination of PM_{10}, based on experiments conducted within the European Committee for standardisation (CEN, 1998). One of these is a high-volume sampler with a PM_{10} inlet, the other is a medium-volume sampler, also with a PM_{10} inlet.

Loss of particles in tubes and connections.

Particles are lost in tubes by deposition to the tube walls, and care should be taken to avoid sharp bends and irregularities. For laminar flows, gravitational settling in horizontal sections will be the main effect, in turbulent flows deposition also occurs in vertical sections. Interestingly, Reynolds numbers of around 2000 seem to give the smallest losses (Ström, 1972). The effects may be serious for particles larger than 3-5 μm a.e.d. and it is generally recommended to avoid tubes and conduits in aerosol sampling.

Collection of particles on filters.

Filter collection efficiencies depend on particle sizes, filter characteristics and flow rates. These samplers were designed for the quantification of suspended particulate matter by subsequent weighing of sample filters. Inlets with 10 μm cut-off are also being used in connection with chemical analyses of collected samples. Maenhaut and co-workers have designed a sampling inlet to be used together with a two-filter filter holder for collection of particles less than 2.5 and between 2.5 and 10 μm. The unit has been tested by Hopke et al.(1997), who found that the collection efficiency follow the guidelines for a PM_{10} sampler.

The penetration of submicron particles through different commercially available filter materials have been studied by Liu and his co-workers (e.g. Liu et al., 1983).

It is noteworthy that fibrous filters, particularly cellulose filters, have considerable penetration for particles around 0.1 μm a.e.d , and that this penetration decreases for increasing flow rates. This is in accordance with filtration theory. The collection efficiencies of Nucleopore filters depend also on flow rate in relation to pore and particle sizes.

Membrane filters have generally good collection efficiencies over a wide range of particle sizes.

Loss and absorption of gases from and to filters and samples collected on filters.

At ambient temperatures and relative humidities, aerosol particles and droplets contain water and other constituents which will be volatilized at higher temperatures and lower relative humidities. The slight pressure drop across the filter may also induce gasification of some species. Further chemical reactions may take place on the filters if the composition of the sample air changes during the sampling period, or simply because particles of different chemical composition are brought into physical contact on the filter substrate. There is no general solution to these problems, although short sampling periods would seem to reduce some of the artefacts.

In general, these sampling problems are related to the contributions of water, ammonium nitrate and volatile organic compounds to the particle mass. Standard procedures for weighing of filters usually specify “conditioning” of the filters at e.g. 25°C and 50% relative humidity for 24 hours before weighing. While this relative humidity is below the deliquescence point for most of the relevant water-soluble salts, it is still not sufficient to remove the water associated with these salts in a real aerosol sample. The procedure does not control the evaporation of any ammonium nitrate or other volatile components from the sample during the conditioning process. The gravimetric determination of PM_{10} is thus operationally defined, and may deviate to some extent from the “true” water-free aerosol mass in the ambient air sampled. The magnitude of this deviation will depend on the chemical composition of the aerosol.

Continuous or semi-continuous monitoring involving either β-ray absorption or modification of the resonance vibrating frequency of piezoelectric crystals or tuning forks require heating of the sampling compartment in order to reduce the contributions of water to the aerosol mass, resulting also in loss of volatile organic components and ammonium nitrate. Again, the PM_{10}, $PM_{2.5}$ or TSP values derived from such measurements are operationally defined, and the results may deviate from gravimetric determination of aerosol mass, or from aerosol mass determinations by other methods. However, there are also cases when the different methods may give quite comparable results. This evidently depends on the chemical composition of the aerosol sampled. The separation of ammonium nitrate in ambient air from gaseous nitric acid and ammonia by diffusion denuders is described by Ferm (1979, 1982), Allegrini et al.(1987), and others.

In addition, to the need for data quality there are requirements for harmonization of measurements. A consideration of sampling deficiencies such as removal of liquid water associated with the particulate phase (Meyer et al., 1995), neutralization of acidic particles (Koutrakis et al., 1988) and loss of volatile particles have to be adequate addressed before certain conclusions from measurements are made. Questions relating to the representativeness of the monitoring site have also to be taken into consideration, as well as, the spatial and temporal variations during the monitoring period. In addition, sampling and analysis has to be representative for human exposure studies and consequently risk assessment evaluations (Willeke and Baron, 1993; Chan and Lippmann, 1980; ACGIH, 1994; Air Quality, 1996; Allen et al., 1997).

Sampling of aerosols is concentrated in measuring their number/mass distribution and in chemical analysis of the different sizes. This is mainly due to the fact that aerosols exist at many different size spectra and arise from multiple sources under different meteorological conditions. Since, the shapes and densities of the atmospheric aerosols are different and also due to the fact that each sampling technique is based on measuring different properties of the aerosols it is necessary to convert the size distribution to an aerodynamic basis distribution. In addition, the sampling methods have to correlate to human inhalation characteristics and to be cost effective and accurate.

In the following different methods of aerosol sampling are reviewed presenting their characteristics, limitations and future developments. The sampling methods reviewed include instruments for large particles (PM_{10}, TSP), fine particle (impactors, cyclones) and for automated sampling (oscillating microbalance, Beta Gauge, Nephelometer).

4.2 Sampling efficiencies and cut-points

An important parameter in aerosol measurements is the adoption of the required cutpoint considerations (U.S. EPA, 1987, Ogden, 1992; U.S. EPA, 1996; Mark and Vincent, 1986). The introduction of the 10 μm (PM_{10}) cutpoint in 1987 (U.S. EPA, 1987) provided a first standardization sampling method which is based on the inhalation model of Lippmann and Chan (1979). However, cutpoint sizes and separation sharpness are not uniformly defined in national and international conventions (ISO, 1993; CEN, 1993; ACGIH, 1994). The different cutpoints can be categorized as Respirable, Thoracic and Inhalable (U.S. EPA, 1996). For example, the thoracic cut point definition by the British Standard EN 841 (CEN, 1993) is that of a cumulative lognormal distribution with a median of 11.64 μm and a geometric standard deviation of 1.5, such as that 50% of aerosols with 10 μm mass aerodynamic diameter are in the thoracic region. Recent considerations of the 2.5 μm cutpoint are mainly concerned on the separation of the fine and coarse mode of ambient particulate matter (U.S. EPA, 1996). This has been done mainly because of health and ecosystem effects are correlated primarily with fine particles that are much smaller than 10 μm.

However, there are many points of representativeness and accuracy during aerosol sampling that someone has to consider such as chemical interactions on the filters, volatilization losses and inlet losses (Marple and Rubow, 1976; Vincent, 1988; Liden and Kenny, 1991; Zhang and McMurry, 1992; Anand et al., 1992).

Since sampling tries to mimic the human inhalation process an attention must be paid also to the fact that human breathing is an anisokinetic sampling process and on the definition of the total inhalable fraction of ambient particles in the construction of aerosol samplers (Mark and Vincent, 1986).

Measurement of the Total Suspended Particulates (TSP) is a common practise using a reference sampling device (Federal Register, 1971). The TSP sampling is efficient method for collecting particles with diameters larger than 40 μm but has limitations such as anisokinetic sampling and wind speed sensitivity (McFarland and Ortiz, 1979; McFarland et al., 1977; Willeke and Baron, 1993).

Sampling of PM_{10} aerosols leaded to the construction of appropriate inlets and samplers that operate under anisokinetic conditions (U.S. EPA, 1987; Ranade et al., 1990; John and Wall, 1983; Wiener et al., 1994). A Sierra-Andersen 321B high volume sampler inlet was considered as a reference method by the U.S: EPA (U.S. EPA, 1987). Early problems of the reference operating inlets due to soiling and particle bounce are considered in currently available inlets such as SA1200 which has oiled surfaces to reduce particle bounce and access screws to make possible cleaning (U. S: EPA, 1996).

Fine particle separation is a more technical and expensive method since requires validation testing, integration in existing sampling instruments and consideration of artefacts in separating coarse and fine particles (Marple et al., 1993; Keeler et al., 1988; Sioutas et al., 1994; Stevens et al., 1993). Problems in separation between the different size fractions increase at high ambient relative humidities since water accumulates in the fine fraction and particle size increases (Keeler et al., 1988). Cyclones are also used in addition to impactors as particle separators (they mimic respiratory deposition) but large biases were reported in the literature due to overloading (Groves et al., 1994; Marple et al., 1993; Hering et al., 1990; Winberry et al., 1993). However, due to their simplicity in use they were used in the “Enhanced Method” for sampling acidic aerosols (U.S. EPA, 1992).

Measurements of the aerosol size distribution are mostly performed with the use of impactors (Marple, 1978; Marple et al., 1993; Koutrakis et al., 1990; Hillamo and Kauppinen, 1991). Problems in sampling efficiency of impactors arise due to particle bounce, overloading and interstage losses (Marple et al., 1993; Wang and John, 1988; Vanderpool et al., 1987).

Cascade impactors are collecting particles using a series of coated substrates including a final stage filter for gravimetric analysis. There are many commercially available impactors such as the Andersen MK1 and MK2 7-stage impactors, the Berner 6-stage and 11-stage impactors and the 10-stage Micro-Orifice Uniform Deposit Impactor (MOUDI) (Hillamo and Kauppinen, 1991; Marple et al., 1993; Marple et al., 1991; Vaughan, 1989; Fang et al., 1991). Calibration is critical during the operation of cascade impactors as several studies showed (Vaughan, 1989; Marple et al., 1991). In addition, long sampling times usually required for chemical analysis or weighting which are also very costly.

4.2.1 Automated Sampling

The need for ambient measurements of particulate matter with a detailed temporal resolution resulted to the development of automated sampling methods. The British smoke shade sampler (BS) uses s 3m plastic tube to deposit ambient particles on white filter paper with the help of an inverted funnel (Bailey and Clayton, 1980; Mage, 1995). Daily samples can be obtained with the BS method with measuring the blackness on the filter. Another similar approach is the Coefficient of Haze automated method in which the particles are collected on a filter-paper that is periodically moved. Opacity measurements determine the concentration of mainly carbon particles deposited (Ingram and Golden, 1973).

A more recent and considerable more advanced automated method is the Tapered Element Oscillating Microbalance (TEOM®) sampler (Patashnick and Rupprecht, 1991). The instrument consists of an oscillating tapered tube with a filter. The measurement method is based on the determination of the shift of oscillating frequency of the tube due to change of the filter mass. The filter performs with a PM_{10} cutpoint inlet and the sampler heats the incoming air to a constant temperature of 30 or 50^oC to maintain moisture in the vapour phase. The mass transducer is also heated to 50^oC to maintain stable measurement conditions.

Biases under certain measurement conditions have been reported in the literature due to volatilization of the aerosol mass sampled in the filter (Hering, 1994; Cahill et al., 1994; Allen, 1998). There is an extensive literature concerning the performance of the TEOM® sampler including the studies of Patashnick and Rupprecht (1991), Patashnick (1998), Cahill et al. (1994), Hering (1994), Allen (1998), WESTAR, 1995).

A method based on a beta attenuation method is also widely used to determine the attenuation of deposited aerosols on a sampler using radioactive sources (Merrifield, 1989; Spagnolo, 1989; Arnold et al., 1992; Wedding and Weigand, 1993). The beta gauge method needs an electronically calibration regression before use in order to give accurate results. Comparison field studies between the beta gauge and other gravimetric methods are reported by Wedding and Weigand (1993) and WESTAR (1995).

Nephelometer is another instrument used to determine the ambient aerosol concentration. The method is based on measuring the light scattering of aerosols integrated under a variety of angles. The instrument measures the scattering coefficient of particles which can be inverted to determine the particle mass based on Mie theory (Hulst, 1981). Overview of the nephelometer applications, limitations and field studies can be found, including the works of Hinds (1982), Gebhart (1993), Rood et al. (1987) and Eldering et al. (1994).

4.2.2 Sampling Needs and Specifications

Geographical representativeness and measurement averaging time are two important factors in sampling methodology. In addition to the need of adequate sampling period to obtain a representative picture of the aerosol temporal variability there is necessary a chemical analysis of the samples to determine the composition/source identification of the particulate matter.

4.2.2.1 Sulphate/Nitrate/Organic

Measurements of sulphate mass is based on continuous (spectroscopic, chemi-luminescent) and time integrated (filter pack, denuders, impactors) sampling (Sioutas and Koutrakis, 1996). Continuous methods are described by Hutzincker et al. (1982). An extensive literature concerning sulphate measurement techniques includes the studies of Koutrakis et al. (1988, 1990), Pierson et al. (1989) and Bennett et al. (1994).

Measurements of particle nitrate includes sampling with filter packs, sampling with filters (nylon or impregnated) operated after a denuder that removes the vapour nitric acid, and also sampling with impactors (Hering et al., 1988; Shaw et al., 1982; Wall et al., 1988; John et al., 1988; Pierson et al., 1988). Inter-comparison between the different methods is performed by Hering et al. (1988), Solomon et al. (1988), Wall et al. (1988) and Pakkanen et al. (1999).

Pakkanen et al. (1988) performed a comprehensive intercomparison study of particulate nitrate and ammonium using various types of filter packs and denuder systems. The virtual impactor collected on average 17% more nitrate than the denuder systems, 24% more than filter packs and 37% more than the Berner low-pressure impactor. The nitrate discrepancy is expected to be mainly due to inefficient coarse particle transport into the denuder systems, fine particle nitrate evaporation in the filter packs and Berner low-pressure impactor.

A review article concerning with measurements of carbonaceous aerosols is written by Hering et al. (1990). Quartz filters, adsorption-corrected quartz filters and impactors were used for ambient sampling of aerosols. Variations between the different sampling methods of carbonaceous aerosols were reported in the literature. Impactor measurements are in general lower than filter measurements by as much as 50% (Chow et al., 1993; Hering et al., 1990).

4.2.2.2 Bioaerosols

There is a variety of techniques used in detecting bioaerosols due to the complexity of the particles involved. In Table 8 we summarize the measurement techniques used in analysis of bioaerosols as presented in the EPA review (1996). A recent review of bioaerosol sampling is given by Mathias-Maser (1998).

Table 8: Analytical methods used in bioaerosol analysis.

	Kinds of Agents	Examples	Sampling Considerations
Culture	Culturable organisms	Fungal spores, yeasts, bacteria, viruses (rarely used)	Viability must be protected
Microscopy	Recognizable particles	Pollen, fungal spores, bacteria	Good optical quality is required
Immunoassay	Agents that stimulate antibodies	Allergens, aflatoxin, glucan	Agents must be elutable from sampling medium.
Bioassay	Agents exerting observable effects in a biological system	Endotoxin, cytotoxins	Same as immunoassay
Chemical assays	Chemicals with recognized characteristics	Trichothecene toxins	Same as immunoassay
Molecular techniques	DNA or RNA-containing particles	Specific organisms	

4.2.2.3 Chemical Composition Analysis

The chemical composition of the ambient particulate matter is a very important component in understanding the pollution sources, chemical reactivity and physicochemical properties of aerosols and their linkage to health effects. Chemical characterization is based in a variety of methods and each of them measures specific chemical species. In accordance with the EPA (1996) review on ambient aerosols we divide the chemical analysis methods into 4 categories :

- Mass
- Water-soluble ions
- Elements
- Organics

Analysis of the chemical species and water does not explain the total aerosol mass but a fraction of it ranging from 65 to 85% from filter samples (EPA, 1996). Besides the main 4 mentioned analytical methods there are other less common analytical methods, among them the isotopic abundances (e.g. Hirose and Sugimura, 1984), mineral compounds (Davis, 1980) and functional groups (Mylonas et al., 1991; Allen et al., 1994).

Mass measured methods include beta attenuation, microbalance methods and gravimetric analysis. In the gravimetric analysis the filter mass is weighted before and after sampling under controlled temperature and relative humidity conditions. Recent studies in this field includes the work of Watson et al. (1989), Feeney et al. (1984), Engelbrecht et al. (1980) and Courtney et al. (1982).

The element analysis include a variety of techniques such as neutron activation analysis (INAA), photon-induced x-ray fluorescence (XRF), particle-induced x-ray emission (PIXE), atomic absorption spectrophotometry (AAS), inductively-coupled plasma with atomic emission spectroscopy (ICP/AES) and scanning electron microscopy with x-ray fluorescence (SEM/XRF). A detailed overview of the various measurement techniques is given by EPA (1996). Peer-reviewed literature concerning the analytical methods for measuring chemical elements in the particulate phase include the works of Hammerle and Pierson (1975), Torok and Van Grieken (1994), Dzubay et al. (1989), Cahill et al. (1989), Cahill and Wakabayashi (1993), Malm et al. (1994), Gordon (1974), Cornille et al. (1990), Turpin et al. (1993, 1994), Hopke and Casuccio (1991) and Schamber (1993).

The wet chemical analysis involves the water-soluble part of atmospheric particulate matter. Ion chromatographic analysis is used for analyzing polyatomic ions such as chloride, nitrate and sulphate. Automated colorimetric analysis is also used for nitrate anions, sulphate and ammonium species. Ions such as soluble sodium, magnesium, potassium and calcium are better measured using atomic absorption spectrometry (AAS). Literature in wet chemical analysis includes the papers of Fung et al. (1979) and Fernandez de la Mora (1989),

Finally, analysis of organic compounds is performed using the glass or quartz-fibre filters that are used during the collection process. Volatilization problems arising because of the high equilibrium vapour pressures of the organic compounds is an important artefact during sampling. In addition, adsorption of gaseous substances on filters or on particles can lead to over-estimation of the organic mass. Bioassay-directed chemical analysis is used for identifying the nonpolar and semi-polar organics in filter samples. However, less work has been devoted to the characterization of the polar organic fraction. The literature concerning analysis of organic particulate matter is very large and increases quickly mainly due to the fact that organics comprise a major fraction of the airborne particulate matter and their importance matter on human health and radiative properties of clouds. Recent studies on sampling of organic matter can be found in the works of Ligocki and Pankow (1989), Bidleman et al. (1986), Tang et al. (1994), Parmar and Grosjean (1990), Arey et al. (1988), Rogge et al. (1993), Kaupp and Umlauf (1992), Rogge et al. (1993), Atkinson et al. (1988), Chow et al. (1993, 1994, 1995), Hering et al. (1990), Van Vaeck et al. (1984), Schwartz et al. (1982) and Hildemann et al. (1991).

4.2.3 Harmonization of Measurements and Air Quality Standards

There is no current standardised method for measuring PM_{10} and $PM_{2.5}$ in Europe but there are recent efforts for PM_{10} standardisation is under development (Position Paper, 1998). Since fine particles are more related to human health (EPA, 1996; Wilson and Suh, 1997) standardisation of $PM_{2.5}$ measurement techniques will be a necessary step in particulate matter measurements.

There is a current legislative effort concerning the development of air quality standards on PM concentrations in Europe (European Commission, 1997; Position Paper, 1998). Until recently the PM standards were controlled under Council Directive 80/779/EEC on Air Quality Limit Values and Guide Values for Sulphur Dioxide and Suspended Particulates (Black Smoke and TSP methods). The limit values for TSP were dependant on the simultaneous concentrations of SO_2. In a recent proposal for a Council Directive relating to limit the value of particulate matter concentrations in ambient air (European Commission, 1997) the proposed limit values for PM_{10} is an annual value of 30 micrograms per cubic meter (which will be reduced to 20 micrograms per cubic meter after 2005) and a daily limit of 50 micrograms per cubic meter. The date by which limit value is to be met is proposed to be the 1 January 2005. The proposed limit values for $PM_{2.5}$ is an annual level of 20 micrograms per cubic meter and a daily value of 40 micrograms per cubic meter. The date by which action level should be met is proposed to be 1 January 2005.

The above Council Directive plans to take into account natural contributions of PM_{10} mainly in the drier parts of the Community. In case that Member States can demonstrate non compliance with the current Council Directive because of high concentrations of natural dusts must adopt action levels for $PM_{2.5}$ (European Commission, 1997).

There is a revision of particulate matter standards in United States which also revises the monitoring requirements for $PM_{2.5}$. The Environmental Protection Agency (EPA) is revising the primary (health-based) PM standards by adding a new annual $PM_{2.5}$ standard set at 15 micrograms per cubic meter and a new 24-hour $PM_{2.5}$ standard at 65 micrograms per cubic meter. EPA is retaining the annual PM_{10} standard of 50 micrograms per cubic meter and adjusting the PM_{10} 24-hour standard of 150 micrograms per cubic meter by changing the form of the standard. The new standard would be met when the 3-year monitored concentrations at the highest monitor in an area is less than or equal to 150 micrograms per cubic meter.

For the moment there are 3 classes of equivalent methods for $PM_{2.5}$ (EPA, 1996). Class I equivalent methods provide capability for collection of several sequential samples automatically without intermediate operator service. This will permit easier and more cost efficient sampling on daily basis. Class II equivalent methods include all other filter methods that produce a 24-hour measurement. Finally, Class III equivalent methods include both continuous or semi-continuous methods.

The EMEP measurement programme does not include particle mass. In principle, this involves only weighing of the aerosol filter in the filterpack used for the determination of sulphate aerosol. Experience indicates that this is also appropriate for the determination of the mass due to ammonium nitrate. However, the simple inlet used for the filterpack sampler may not satisfy the criteria for determining particle mass, in particular not aerosol mass below 10 μm or below 2.5 μm aerodynamically equivalent diameters, respectively. The influence of particles larger than 10 μm on the weight of the sample filter can be disregarded, as wind tunnel and other experiments have shown that the uptake of particles larger than 5-7 μm is reduced, particularly under windy conditions. A modified air inlet should therefore be used, if filters are to be weighed.

Separation of aerosols according to size is another technical problem. Cascade impactors with improved performance are now readily available, but the pressure drop in the last few impactor stages used to collect particles less than 1 μm will cause the volatile constituents, ammonium nitrate and some organic compounds to disappear. These constituents must then be assessed separately. It is also possible to determine the aerosol size distribution by other means, e.g. by optical classifycation and counting of individual aerosol particles. However, these instruments are expensive to purchase and maintain.

A separation between particles of aerodynamic diameter less than and larger than 2.5 μm is usually called for in air quality monitoring. This is achieved in instruments like the EPA-commissioned dichotomous particle sampler, which uses a virtual impactor step to separate 90% of the fine particle fraction from the larger particles. Both fractions are then collected on filters for subsequent weighing of the filters, or chemical analyses. A membrane filter with uniform pores, the so-called Nucleopore filter, can also be used to collect large particles by impaction when the filter is coated with an appropriate sticky material. While this separation principle is somewhat more subject to artefacts, the simplicity of operation and practical experience suggests that the use of two-stage aerosol filters will give useful information when combined with the EMEP filter pack system.

5 Emissions and Sources of Atmospheric Particles

The sources and emissions of particulate matter is a more complex process compared to the specific gaseous pollutants. This is due to differences in the emission characteristics (chemical composition/size distribution) of emitted particles from a variety of natural and anthropogenic sources. In Table 9 and Table 10 we show a comprehensive overview of the major sources of particulate matter as presented in the U.S. Environmental Protection Agency review (1996).

The TNO Institute of Environmental Sciences, Energy Research and Process Innovation in the Netherlands have compiled an emission inventory for PM_{10}, $PM_{2.5}$ and $PM_{0.1}$ in Europe for the years 1990 to 1993 (TNO, 1997). The 1990 data were spatially distributed on a 0.5° x 1°.

The data summarized for 26 European Countries for 1993 are shown in Appendix 1. From the Tables in Appendix 1 we can conclude that the per capita emissions are in general higher in Central and Eastern European Countries (CEE) compared to the Western European Countries. The former USSR Countries have even higher emission rates. Expected reasons for these differences are due to the greater use of solid fuels for stationary combustion in the CEE countries and former USSR and lack of effective emission abatement measures (Airborne Particles Expert Group – Interim Report, 1998). From the Tables it can be seen a fall in the emissions per capita in Western Europe and an increase in Central and Eastern Europe. In Table 11 we present the particulate matter emissions in Germany (Position Paper, 1998). In Appendix 2 we summarize available data on size distribution/chemical composition of emitted particles from different sources (Kumar and Lurmann, 1996).

A comprehensive summary of the particulate matter emissions in Europe can be found in the reports by TNO (1997), Position Paper (1998) and Airborne Particles Expert Group – Interim Report (1998).

Table 9: Constituents of atmospheric fine particles (< 2.5 µm) and their major sources (EPA, 1996).

Sources				
Primary			Secondary	
Aerosol species	Natural	Anthropogenic	Natural	Anthropogenic
SO4 (=)	Sea spray	Fossil fuel combustion	Oxidation of reduced sulphur gases emitted by the oceans and wetlands; and SO_2 and H_2S emitted by volcanoes and forest fires.	Oxidation of SO_2 Emitted from fossil fuel combustion.
NO3 (-)	--	Motor vehicle Exhaust, large combustion sources	Oxidation of NO_x produced by soils, forest fires and lighting	Oxidation of NO_x emitted from fossil fuel combustion and in motor vehicle exhaust.
Minerals	Erosion, Re-entrainment	Fugitive dust; paved, unpaved roads; agriculture and forestry	--	--
NH4(+)	--	Motor vehicle exhaust	Emissions of NH_3 from wild animals, undisturbed soil	Emissions of NH_3 from animal husbandry, sewage, fertilized land
Organic Carbon	Wild fires	Open burning, wood burning, cooking, motor vehicle exhaust, tire wear	Oxidation of hydrocarbons emitted by vegetation, (terpenes, waxes); wild fires	Oxidation of hydrocarbons emitted by motor vehicles, open burning, wood burning
Elemental Carbon	Wild fires	Motor vehicle exhaust, wood burning, cooking	--	--
Metals	Volcanic activity	Fossil fuel Combustion, smelting, brake wear	--	--
Bioaerosols	Viruses, bacteria	--	--	--

Table 10: Constituents of atmospheric coarse particles (> 2.5 µm) and their major sources (EPA, 1996).

Sources				
	Primary		Secondary	
Aerosol species	Natural	Anthropogenic	Natural	Anthropogenic
Minerals	Erosion, Re-entrainment	Fugitive dust, paved, unpaved road dust, agricultural and forestry	--	--
Metals	Erosion, re-entrainment, organic debris	--	--	--
Miscellaneous ions	Sea spray.	Road salting	--	--
Organic carbon	--	Tire and asphalt wear	--	--
Organic debris	Plant, insect fragments	--	--	--
Bioaerosols	Pollen, fungal spores, bacterial agglomerates	--	--	--

Table 11: Particulate matter emissions and major sources in Germany (Position Paper, 1998).

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

6 Concentration of Particulate Matter in Europe

6.1 Overview

The industrial activity, the high volumes of traffic and the urbanization of the European Continent had as a result the anthropogenic sources of PM to be predominant, mainly in urban areas (Position Paper, 1998). Particulate Matter can be of primary origin (directly emitted) or secondary formed in the atmosphere from vapour precursors (Seinfeld and Pandis, 1998). Fine particles can be transported over long distances. Sahara dust (Rahn et al., 1979) and volcano (Haulet et al. 1977; Malinconico, 1979) emissions are two main sources of primary particles in Europe. Other natural sources include biological particles (Matthias-Maser and Jaenicke, 1995), sea spray (Savoie and Prospero, 1982), and soil resuspended by the wind (Nicholson, 1988). Anthropogenic sources in Europe are similar and include combustion sources (residential and industrial), traffic, power plant emissions, industrial fugitive dust, man-made forest fires and various other human activities such as building construction.

Secondary particles can be arise from natural sources such as secondary organic particles formed from vegetation, natural particulate sulphate from sea phytoplankton and ammonia component in PM. Secondary aerosols comprise a major part of the fine ambient aerosol mass and because they reside mainly in the accumulation mode they can be easily inhaled and reach the lower human respiratory tract. Literature concentrated with ambient aerosol concentrations include the works of Berdowski et al. (1998), European Commission (1996, 1997), EMEP-WMO (1997), Kitto and Harisson (1992), Lee et al. (1994), Mihalopoulos et al. (1997), Millán et al. (1997), McDowell et al. (1990), Ritter (1997), Quality of Urban Air Review Group (1996), Simpson et al. (1996), WHO (1987) and Ziomas et al. (1998).

6.2 Global and Continental Scale Aerosol Pattern

6.2.1 Long Range Dust Transport

Airmasses of Sahara dust can be transported from North Africa over long distances into the NE Atlantic and in Europe (Diouri et al., 1998). The amount of Saharan material deposited in the north-western Mediterranean is estimated to be close to 3.9 million of tonnes every year (Loyle-Pilot et al., 1986). Contributions to Northern part of Europe from the Sahara long-range transport is also reported (Junge, 1972).

At the moment the exact percentage contribution of the Saharan dust to PM_{10} concentration in Mediterranean is not fully resolved (Position Paper, 1998). Saharan dust is composed of quartz calcite, gypsum, and clay minerals. (Roda et al., 1993).

6.2.2 Volcano Emissions

Volcanoes are an important source of PM_{10} and SO_2 (which can serve to produce secondary particles). In Europe there is still volcanic activity at the Canaries, Iceland and Sicily. In Italy there is an average emission of 4,000 tonnes/day SO_2 from mountain Etna, which definitely has an effect on the composition of particulate matter in the local region (Haulet et al., 1977).

6.3 European Aerosol Patterns and Trends

Particulate Matter measurements are insufficient to have a complete picture of PM concentrations across Europe. Measurements of PM are concentrated in urban areas and in addition there is no sufficient reporting of the data on international organizations (Europe's Environment, 1995). However, the ambient concentration of particulate matter at a defined spatial and temporal point is a complex functional of emissions, dispersion, physico-chemical processes and removal. The aerosol residence time is an important factor that determines the ambient aerosol concentration. Fine aerosols in the size range of 0.1–1.0 µm have residence time of several days and if they are transported in the upper troposphere their residence time increases to several weeks.

Measurements of PM_{10} and black smoke (BS) wintertime (winter 1993/1994) concentrations in Europe during the PEACE study (Hoek et al., 1996, 1997) show small differences between urban and non-urban locations. Similar winter studies for PM_{10}, $PM_{2.5}$ and BS in the Netherlands (winters of 1992/1993, 1993/1994 and 1994/1995) show the same trend in the results (Van Der Zee et al., 1998). PM_{10} and BS concentrations were on average 13% and 19% higher in the urban areas than in the non-urban areas. Non-organic secondary aerosol concentrations were on average 8% lower in the urban areas whereas $PM_{2.5}$ concentrations were similar in between urban and non-urban sites. Also higher elemental concentrations in PM_{10} were found in the urban area for all elements except Si. Meteorological conditions seem to play a crucial role in the observations. Easterly winds resulted to smaller differences between urban and nonurban sites showing the importance of the regional aerosol component from Eastern Europe whereas winds from the sea lead to lower average PM concentrations and larger differences between urban and nonurban locations.

Measurements of PM_{10} have been also performed in Switzerland during 1993 at a dozen urban, rural and alpine sites. The PM_{10} concentrations ranged between 10 $\mu g\ m^{-3}$ (alpine region) to 33 $\mu g\ m^{-3}$ (urban site). An interesting observation is that the rural levels were one-third than the urban levels and homogeneously distributed (Monn et al., 1995). In the Alpine sites the levels were considerably lower. Aircraft measurements over the Swiss plateau during photosmog episodes were also performed and showed that under conditions of strong air pollution and strong photochemical activity the aerosol particles grow with secondary processes (such as condensation) into the diameter range 0.12-1.5 µm (Hering et al., 1998).

Monn et al. (1995) found also that in Switzerland the ratio between PM_{10} and TSP to be between 0.6 and 0.75 with the highest ratios in the urban sites. Seasonally there were higher concentrations of PM during winter and autumn due to cold temperature-inversions.

Sampling of aerosols in an EMEP station Iskrba in Slovenia has occurred in spring 1997 and winter 1998 using a low pressure Berner impactor (Tursic et al., 1998). Major components include sulphate, organic and elemental carbon, nitrate and chloride. The highest concentrations of SO_4^{4-} and NH_4^+ are in the accumulation mode, while in the coarser fraction there are high concentrations of Na^+, Mg^{2+}, Ca^{2+} and heavy metals. It was not observed a diurnal black carbon cycle as in an urban site and the concentration of PM was higher in spring than in the winter.

Long-range transport of particulate matter has been also studied intensively in southern Scandinavia (Pakkanen et al., 1996; Lannefors et al., 1980,1983; Martinsson et al., 1984; Oblad and Selin, 1986; Amundsen et al., 1987, 1992; Cornille et al., 1990). Air masses originating from the British Isles, central and eastern Europe were mainly responsible for the long range transport of pollution such as sulphur. A long-range transport model has been developed to in the EMEP framework to study the impact of sulphur pollution in Scandinavia and in Europe in general (Eliassen and Saltbones, 1983). During long-range transport episodes, higher concentrations of pollutants by 20-50 times compared to the background levels at Birkenes site in Southern Norway (Pakkanen et al., 1996). Measurements of the aerosol size distribution using a Berner impactor showed one or two modes in both the fine mode and coarse modes. The use of a stacked filter unit (SFU) and low-pressure impactors (LPIs) in two locations in Norway (Birkenes and Nordmoen) showed similar average values for elemental fine to total aerosol ratios. Furthermore, the elemental ratios for several pollutant elements of the air masses in southern Norway (Pakkanen et al., 1996) and southern Sweden (Swietlicki et al., 1989) are very similar.

Biological aerosols may also have a considerable influence in the mass size distribution and studies in an urban/rural influenced region in Mainz indicated that biological aerosols can constitute close to 30% of the total aerosol mass (Matthias-Maser and Jaenicke, 1995). These results are valid for an urban/rural influenced region. Results from measurements on the Atlantic Ocean show a percentage of biological aerosols to total aerosol of only 14% in number and 8% in volume (Matthias-Maser, 1998).

6.3.1 Sea Spray

Sea spray aerosols have the same composition as seawater enriched in organic matter (marine-derived sterols, fatty alcohols, and fatty acids) that are present at the ocean surface (Seinfeld and Pandis, 1998; Schneider and Gagosia, 1985). Sea sprays are mostly produced from the breaking waves on the sea surface and have a size range between 1–20 μm (Blanchard and Woodcock, 1980) and the smallest sea particles can have a lifetime of three days Junge, 1972). Chemical reactions on the surface of sea-salt particles may lead to a “chloride deficit” in the marine aerosols such as the reaction of sulphuric acid or nitric acid with sodium chloride which will produce gaseous hydrochloric acid and sodium sulphate or nitrate; respectively.

It is clear that coastal regions and islands are more affected by the presence of sea-sprays but sea particles have been measured in inland regions. In the northern Europe the use of de-icing salt on the roads will also result in increased concentration levels of sodium chloride, mainly in urban areas and close to the roads (Position Paper, 1998).

6.3.2 Composition of Nonurban Aerosol

A chemical mass balance of fine particles (diameter smaller than 1.5 μm) collected at three European sites was reported recently (Zappoli et al., 1998). The sites were located in southern Sweden (S), in a rural area in the Great Hungarian Plain (H) and in the polluted Po Valley, northern Italy (I). The total fine aerosol mass ranged from a value of 38 μg m^{-3} (I) in the most polluted area to around 6 μg m^{-3} (S) in the background site. In rural station in Hungary showed a value close to 23 μg m^{-3} (H). Speciation of the aerosol components with reference to water solubility of the different species is presented in Table 13. The results show a very high water solubility and higher fraction of WSOC in the background site compared to the rural and polluted sites (Zappoli et al., 1998).

Table 12: Speciation (in % units) of different aerosol classes of components EC=elemental carbon, WISOC=water-insoluble organics, WSOC=water-soluble organics, OSII=other soluble inorganic ions, WISIC=water-insoluble inorganics, ND=non detected (Zappoli et al., 1998).

site	EC	WISOC	WSOC	OSII	NH_4^+	SO_4^{4-}	NO_3^-	WISIC	ND
S	1	7	36	11	4	20	4	15	-
H	2	13	14	2	10	32	8	n. a.	18
I	3	7	14	14	10	20	16	11	6

6.3.3 Urban European Aerosol Pattern

The concentration of urban aerosols is a mixture of emissions from industry, traffic, various combustion sources, secondary formation, natural sources and from long range transport. The number size distribution is dominated by fine particles, whereas the mass size distribution has two modes, the accumulation and the coarse modes (Seinfeld and Pandis, 1998).

There are recent attempts for monitoring of urban air quality in the European cities with the establishment of the European-wise air quality monitoring and information network of the European Environment Agency (EEA) which is called EUROAIRNET (EEA, 1997; Larssen and Lazaridis, 1998). Until now there have selected over 800 stations in Europe which monitor various gaseous pollutants, as well as, TSP, PM_{10}, $PM_{2.5}$ and Black Smoke. Literature concerning monitoring and levels of pollutants in urban areas in Europe include the reviews by van Aalst et al. (1998), Larssen and Lazaridis (1998), CEC (1997), EEA (1997) and WHO (1997).

In UK the urban sulphate concentrations are similar to rural levels. The rural to urban ratio for nitrate varies from 0.46 – 0.84 showing therefore an urban source of nitrate measures (Airborne Particles Expert Group – Interim Report, 1998) (see also Table 13). The mass median diameters of sulphate, nitrate and ammonium measured in Leeds (UK) were 1.1 μm, 1.6 μm and 1.0 μm respectively. Model calculations show that both UK and continental European sources of sulphur dioxide contribute to sulphate pollution episodes in the UK measures (Airborne Particles Expert Group – Interim Report, 1998). An overview of the urban patterns in Europe can be found in the Position Paper (1998). In the United States there is currently comprehensive database for particulate matter and various gaseous pollutants through the internet (AIRS, 1995).

Table 13: Weekly average mass concentrations of aerosol component at Leeds 1995/6 ($\mu g\ m^{-3}$) measures (Airborne Particles Expert Group – Interim Report, 1998).

Week beg.	Urban NO_3^-	Rural NO_3^-	Urban SO_4^{2-}	Rural SO_4^{2-}	Urban NH_4^+	Rural NH_4^+	Total Mass	Sum of Salts*
15-May-95	1.16		2.32				18	4.7
21-Jun-95	4.13		3.73				26	10.5
05-Jul-95	4.53		5.48				54	13.4
26-Jul-95	7.2		6.07				55	17.7
09-Aug-95	2.95	2.42	6.68	5.87	1.18	0.83	42	13.0
18-Oct-95	4.35	2.31	4.34	4.12	0.67	0.36	42	11.6
15-Nov-95	2.77	1.27	3.96	2.80	1.22	0.94	44	9.0
17-Jan-96	5.26	4.43	11.18	9.57	4.89	2.35	69	22.2
Average	4.04	-	5.47	-	-	-	44	12.7

* Sum of ammonium sulphate and nitrate: 1.29 NO_3^- + 1.375 SO_4^{2-}. Also NH_4NO_3 = 1.29 x NO_3 and $(NH_4)_2SO_4$ = 1.375 x SO_4.

There is a vast literature concerning patterns and trends of ambient aerosols and their concentration characteristics. The interested reader can be referred to the reviews by Jaenicke (1993), Pruppacher and Klett (1997), Seinfeld and Pandis (1998), Position Paper (1998) and EPA (1996).

7 Human Exposure and Health Effects of Atmospheric Particles

In recent years, extensive research effort has been invested in examining the relationship between exposure to particulates and resulting health effects. Data from epidemiological studies conducted to date demonstrate associations between ambient particulate concentrations and increased morbidity and mortality, while data from toxicological studies have begun to provide potential biological explanations for these observed associations. Morbidity and mortality and their association with particulate matter exposures occur in fact below the current air quality standards (EPA, 1996a).

The characteristics and potential effects of large or “coarse” (mass median aerodynamic diameter between 2.5 and 10 microns) and fine fraction particles (mass median aerodynamic diameter below 2.5 microns) are quite different. Coarse particles are originated mainly from desert dust, agricultural fields, resuspension of dust from roads and sea sprays. Coarse particles can accumulate in the upper parts of the human respiratory tract and lead to health problems such as asthma.

Fine particles are emitted from industrial and residential combustion, vehicle exhaust and are also formed due to secondary conversion from the gaseous to particle phase (from gases such as sulphur dioxide, nitrogen oxides, and volatile organic compounds). Fine particles can penetrate deeply into the alveolar region of the human respiratory system and are more likely to lead to severe health problems than coarse particles (e.g., premature mortality and hospital admissions).

Possible Health Effects due to fine particles mostly *(based on United States Environmental Protection Agency Guidelines)*: Premature death, Respiratory related hospital admissions and emergency room visits, Aggravated asthma, Acute respiratory symptoms, including aggravated coughing and difficult or painful breathing, Chronic bronchitis, Decreased lung function that can be experienced as shortness of breath and Work and school absences.

Risk Groups from exposure to fine particles: The Elderly, Individuals with Pre-existing Heart or Lung Disease, Children, Asthmatics and Asthmatic Children.

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Emissions of particulate matter from vehicles arise due to direct emissions from vehicle exhausts, tyre and brake wear and resuspension of road dust. Emissions from road transport are major source of PM_{10}. Diesel engines 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. Hydrocarbons and sulphates condense later on airborne particles, mainly on the freshly emitted carbon. Some of these compounds are carcinogens and mutagens. 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 European Centre for Environment and Health of the WHO (WHO-ECEH) has been updating the air quality guidelines published first in 1987 (WHO 1987, 1994 and 1996). The current state of knowledge concerning the health effects of these compounds, including their mechanisms of action, has been evaluated and the air quality guidelines have been recommended with the aim to protect populations from the adverse effects of the pollution (WHO, 1996). The review of the WHO serves as an excellent starting point. Besides identification of the hazardous substances in the air and quantification of population exposure, public health assessment needs information about the extent of health response associated with a certain exposure level (Harrison, 1994; Möller et al., 1994). In evaluating the benefits of selective pollution abatement programs and technology, it is important to describe not only the changes to ambient pollution concentrations resulting from the measures but also to assess the improvements to public health.

Major literature reviews in the area of health effects due to PM ambient concentrations are given by Brunekreef et al. (1995; 1998), Dockery, Pope (1994), Institute for Environment and Health (1996), Clench-Aas and Krzyzanowski (1996) Holgate and Anderson (1995) Holgate and Waller (1995). Studies chosen will conform to the recommendations in the COST series of air pollution epidemiology manuals. (COST 613/2, 1994, 1995, and 1997).

There is a need in future exposure studies to consider both acute and chronic health effects. Each type of study provides its own valuable information but has its own limitations[2]:

1 **Animal investigations**

2 Value: imperative in the search for mechanisms of action,

3 Limitation - transferability to the human organism is always a question.

4 **Clinical/Chamber investigations -**

5 Value: imperative in the search for mechanisms of action, directly applicable to the human situation.

6 Limitation - applies only to short-term effects, and does not indicate representatively to the real life situation with simultaneous exposure to other compounds and other meteorological situations.

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[2] Part of the current text on Human Exposure and Health Effects of Atmospheric Particles was written by Dr. Clench-Aas (NILU).

7 Epidemiology/population investigations

8 Value - provides real life response, accounting for simultaneous exposure to other compounds and other meteorological phenomena.

9 Limitation - difficult to isolate the effects to a particular source/a particular compound, size of particle, or to a specified composition.

The health effects of diesel exhaust have been public concern for many years. A summary of some of the major findings is repeated here (Yu and Yoon, 1989). Long term exposure of animals to high concentrations of inhaled diesel exhaust conducted at different laboratories showed that there was an accumulation of diesel soot (Chan et al., 1981, 1984; Griffis et al., 1983; Heinrich et al., 1986; Wolff et al., 1987; Strom et al., 1987, 1988), formation of DNA adducts (Wolff et al., 1986) and incidence of tumours (Brightwell et al., 1986; Ishinishi et al., 1986; Iwai et al., 1986; Stober et al., 1986; Mauderly et al., 1987) in the lungs of rats. No carcinogenic effects of diesel exhaust were observed in hamsters (Cross et al., 1978; Heinrich et al., 1982, 1986) while conflicting results were reported on mice (Campbell et al., 1981; Orthoefer et al., 1981; Pepelko, 1982; Heinrich et al., 1986). Epidemiological studies are generally consistent in observing a small to moderate excess relative risk of lung cancer in workers who were exposed to emissions from older, uncontrolled diesel engines (HEI, 1995).

An overview of recent studies and findings concerning with health effects of PM on humans can be found in HEI (1995), EPA (1992, 1995, 1996a,b), ACEA/EUROPIA (1995), APHEA/PEACE (1995), APHEA (1996), Bjerkens-Haugen et al. (1995), Clench-Aas and Larsen (1993), Clench-Aas et al. (1993, 1998), Dockery et al. (1993), Gordin et al. (1996), Hefflin et al. (1994), HMSO (1995), Ho and Winer (1998), Jantunen et al. (1997), Katsoyanni (1996), Katsoyanni et al. (1995), Kolbenstvedt et al. (1991), Larssen et al. (1993), Larssen (1987), Lazaridis and Georgopoulos (1997), Lawther et al. (1970), Lipfert and Wyzga (1995), NTIS (1997), NRDC (1996), O’Riordan (1989), Pope et al. (1992, 1995), Ross et al. (1998), RIVM/NILU (1995), Schlesinger (1995), Schwartz (1994), Schwartz and Dockery (1992a,b), Seaton et al. (1995), Thurston (1996), Torp and Larssen (1996), Wilson and Suh (1997), Wichman and Heinrich (1995) and WHO (1995).

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8.2 Monitoring, Sampling and Analysis of Particulate Matter

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Appendix A

Table A.1: European inventory for PM_{10} in 1993 in ktonnes. Data from TNO (1997).

Country	Power generation	Industrial combustion	Small combustion	Processes	Road transport	Other transport	Agriculture	Waste processing	**Total**
Albania	0.5	4.5	0.9	0.8	1.3	0.0	0.4	0.0	**8**
Austria	1.3	2.4	9.7	7.5	10.0	1.0	9.1	0.0	**41**
Belgium	1.6	7.4	14.0	21.0	21.0	1.0	17.0	0.7	**84**
Bulgaria	76.0	14.0	7.5	24.0	11.0	1.0	8.8	0.0	**140**
Switzerland	0.1	1.2	12.0	2.3	5.2	0.0	4.3	0.3	**25**
Former Czechoslovakia	140.0	150.0	110.0	64.0	35.0	1.0	18.0	0.0	**520**
Former GDR	250.0	0.4	44.0	39.0	37.0	0.0	9.1	0.0	**380**
Former Western Germany	35.0	79.0	19.0	80.0	160.0	0.0	56.0	0.7	**430**
Germany	285.0	79.4	63.0	119.0	197.0	0.0	65.1	0.7	**810**
Denmark	3.1	2.1	9.7	3.4	11.0	1.0	25.0	0.2	**56**
Spain	14.0	18.0	12.0	30.0	65.0	6.0	48.0	1.5	**190**
Finland	2.0	4.1	5.7	6.5	12.0	0.0	3.3	0.0	**34**
France	1.5	40.0	54.0	49.0	120.0	0.0	53.0	78.0	**400**
UK	21.0	32.0	41.0	44.0	99.0	1.0	31.0	2.8	**280**
Greece	12.0	5.1	6.5	6.4	20.0	2.0	4.7	0.0	**57**
Hungary	31.0	3.9	16.0	21.0	17.0	0.0	22.0	0.0	**110**
Ireland	1.7	1.2	8.7	1.3	5.7	0.3	4.1	0.0	**23**
Italy	35.0	26.0	23.0	49.0	110.0	0.0	35.0	16.0	**290**
Luxembourg	0.0	0.9	0.7	3.5	1.1	0.0	0.0	0.0	**6**
Norway	0.0	3.2	1.3	6.9	5.2	2.5	1.9	0.1	**21**
Poland	510.0	210.0	150.0	85.0	38.0	3.0	45.0	0.0	**1000**
Portugal	4.4	3.1	2.1	5.5	12.0	0.0	8.0	0.0	**35**
Romania	86.0	29.0	6.2	66.0	14.0	1.0	25.0	0.0	**230**
Former USSR (European part)	740.0	950.0	1900.0	1300.0	210.0	100.0	170.0	0.1	**5400**
Sweden	0.6	4.6	6.3	8.9	9.3	0.5	5.9	0.2	**36**
Former Yugoslavia	170.0	26.0	37.0	35.0	15.0	0.0	18.0	0.0	**300**
Europe (Exc. Former USSR)	**1400**	**670**	**600**	**660**	**830**	**30**	**450**	**100**	**4800**

Table A.2: European inventory for $PM_{2.5}$ in 1993 in ktonnes. Data from TNO (1997).

Country	Power generation	Industrial combustion	Small combustion	Processes	Road transport	Other transport	Agriculture	Waste processing	**Total**
Albania	0.4	2.0	0.6	0.3	0.9	0.0	0.2	0.0	**4**
Austria	0.8	1.5	5.8	3.9	6.8	0.4	4.4	0.0	**24**
Belgium	0.9	3.9	7.9	13.0	14.0	1.0	8.3	0.5	**49**
Bulgaria	47.0	6.3	3.5	16.0	7.5	0.2	3.9	0.0	**85**
Switzerland	0.1	0.7	7.2	1.1	3.4	0.0	2.0	0.2	**15**
Former Czechoslovakia	89.0	64.0	47.0	41.0	24.0	0.0	8.7	0.0	**270**
Former GDR	160.0	0.2	18.0	27.0	27.0	0.0	4.4	0.0	**240**
Former Western Germany	22.0	38.0	8.8	48.0	100.0	10.0	27.0	0.5	**250**
Germany	182.0	38.2	26.8	75.0	127.0	10.0	31.4	0.5	**490**
Denmark	1.7	1.2	7.3	1.7	7.3	0.7	12.0	0.2	**33**
Spain	8.7	8.8	7.1	16.0	43.0	5.0	23.0	1.0	**110**
Finland	1.2	2.4	3.6	4.0	7.9	0.4	1.6	0.0	**21**
France	0.9	20.0	34.0	27.0	80.0	2.0	22.0	54.0	**240**
UK	12.0	16.0	21.0	26.0	65.0	5.0	13.0	1.9	**160**
Greece	7.7	2.7	4.2	2.7	13.0	2.0	2.0	0.0	**34**
Hungary	20.0	1.8	7.6	15.0	11.0	1.0	9.2	0.0	**65**
Ireland	1.1	0.7	4.8	0.5	3.8	0.2	1.9	0.0	**13**
Italy	23.0	13.0	15.0	27.0	70.0	1.0	15.0	11.0	**170**
Luxembourg	0.0	0.5	0.4	2.2	0.7	0.0	0.0	0.0	**4**
Norway	0.0	1.7	0.8	4.1	3.4	2.3	0.9	0.0	**13**
Poland	300.0	95.0	69.0	50.0	25.0	2.0	22.0	0.0	**560**
Portugal	2.7	1.9	1.6	2.2	8.1	0.1	3.6	0.0	**20**
Romania	58.0	12.0	3.6	42.0	9.3	0.7	12.0	0.0	**140**
Former USSR (European part)	440.0	430.0	900.0	830.0	140.0	70.0	77.0	0.1	**2900**
Sweden	0.4	2.8	3.7	5.7	6.1	0.5	2.8	0.2	**22**
Former Yugoslavia	110.0	11.0	17.0	24.0	10.0	0.0	8.4	0.0	**180**
Europe (Exc. Former USSR)	**870**	**310**	**300**	**400**	**550**	**30**	**210**	**70**	**2700**

Table A.3: European inventory for $PM_{0.1}$ in 1993 in ktonnes. Data from TNO (1997).

Country	Power generation	Industrial combustion	Small combustion	Processes	Road transport	Other transport	Agriculture	Waste processing	**Total**
Albania	0.0	0.1	0.0	0.1	0.3	0.0	0.0	0.0	**0.6**
Austria	0.1	0.2	0.7	1.0	2.0	0.2	0.0	0.0	**4.1**
Belgium	0.1	0.4	1.1	3.5	4.1	0.4	0.0	0.1	**9.7**
Bulgaria	3.2	0.5	0.3	3.3	2.2	0.1	0.0	0.0	**9.4**
Switzerland	0.0	0.1	1.2	0.2	0.9	0.0	0.0	0.0	**2.5**
Former Czechoslovakia	4.5	3.3	2.7	9.0	7.6	0.0	0.0	0.0	**27.0**
Former GDR	6.8	0.0	0.9	6.0	8.1	0.0	0.0	0.0	**22.0**
Former Western Germany	1.4	2.1	0.5	12.0	29.0	1.0	0.0	0.1	**47.0**
Germany	8.2	2.1	1.4	18.0	37.1	1.0	0.0	0.1	**69.0**
Denmark	0.3	0.1	0.8	0.4	2.1	0.3	0.0	0.0	**4.0**
Spain	0.8	0.9	1.0	3.7	13.0	2.0	0.0	0.2	**22.0**
Finland	0.1	0.2	0.6	1.2	2.2	0.2	0.0	0.0	**4.5**
France	0.1	1.4	4.4	7.5	24.0	1.0	0.0	12.0	**50.0**
UK	1.8	1.3	1.8	7.0	18.0	2.0	0.0	0.4	**33.0**
Greece	0.4	0.2	0.6	0.6	4.1	0.7	0.0	0.0	**6.7**
Hungary	0.9	0.1	0.6	3.8	3.7	0.1	0.0	0.0	**9.2**
Ireland	0.1	0.1	0.4	0.1	1.1	0.1	0.0	0.0	**1.9**
Italy	3.5	1.1	2.1	6.9	21.0	1.0	0.0	2.5	**38.0**
Luxembourg	0.0	0.0	0.1	0.8	0.2	0.0	0.0	0.0	**1.1**
Norway	0.0	0.1	0.1	1.2	1.0	1.1	0.0	0.0	**3.6**
Poland	32.0	4.4	3.4	9.4	7.8	0.3	0.0	0.0	**57.0**
Portugal	0.4	0.2	0.2	0.6	2.5	0.1	0.0	0.0	**4.0**
Romania	4.7	0.7	0.3	8.2	2.9	0.5	0.0	0.0	**17.0**
Former USSR (European part)	66.0	21.0	54.0	150.0	37.0	24.0	0.0	0.0	**350.0**
Sweden	0.1	0.3	0.6	1.6	1.8	0.2	0.0	0.0	**4.6**
Former Yugoslavia	4.0	0.8	1.2	5.1	3.1	0.1	0.0	0.0	**14.0**
Europe (Exc. Former USSR)	**65**	**19**	**26**	**93**	**160**	**10**	**0**	**16**	**390**

Appendix B

Table B.1: PM emissions size distribution data (Kumar and Lurmann, 1996).

Profile Name	$PM_{< 0.625}$ Fraction	$PM_{0.625\text{-}2.5}$ Fraction	$PM_{2.5-10}$ Fraction	$PM_{> 10}$ Fraction
Liquid Material Combustion	0.7907	0.1784	0.0067	0.0242
Fuel Combustion – Residual	0.5542	0.2097	0.1002	0.1359
Fuel Combustion – Distillate	0.7907	0.1784	0.0067	0.0242
Utility Boilers-Residual	0.7816	0.1745	0.013	0.0309
Stat. I. C. Engine – Liquid Fuel	0.7907	0.1784	0.0067	0.0242
Stat. I. C. Engine – Gasoline	0.8058	0.1806	0.0075	0.0061
Stat. I. C. Engine – Diesel	0.7697	0.1711	0.018	0.0412
Vehicular Sources – Gasoline	0.8058	0.1805	0.0075	0.0062
Vehicular Sources – Diesel	0.7696	0.1714	0.0179	0.0411
Marine Vessels – Liquid Fuel	0.7697	0.1711	0.018	0.0412
Gaseous Material Combustion	0.8111	0.1812	0.0077	0
Residential – Natural Gas	0.8111	0.1812	0.0077	0
Stat. I. C. Engine – Gas	0.8058	0.1806	0.0075	0.0061
Petroleum Heaters – Gas	0.7629	0.1707	0.015	0.0514
Solid Material Combustion	0.6403	0.2902	0.0658	0.0037
Coal/Coke Combustion	0.0167	0.14	0.2203	0.623
Stat. I. C. Engine – Solid Fuel	0.6403	0.2902	0.0658	0.0037
Wood Waste Combustion	0.6403	0.2902	0.0658	0.0037
Other Waste Combustion	0.6403	0.2902	0.0658	0.0037
Planned/Unplannd Forest Fires	0.6919	0.1627	0.0227	0.1227
Agricultural Burning	0.6919	0.1627	0.0227	0.1227
Unplanned Structural Fires	0.7165	0.2002	0.0619	0.0214
Fireplaces	0.7162	0.1578	0.0431	0.0829
Aircraft – Jet Fuel	0.0253	0	0	0.9747
Orchard Heaters	0.7907	0.1784	0.0067	0.0242
Incineration – Liquid Fuel	0.7907	0.1784	0.0067	0.0242
Incineration – Gaseous Fuel	0.8111	0.1812	0.0077	0
Incineration – Solid Fuel	0.11	0.103	0.0719	0.7151
Evaporation	0.7617	0.1679	0.0289	0.0415
Coating Material Evaporation	0.7617	0.1679	0.0289	0.0415
Paint Application – Oil Based	0.7617	0.1679	0.0289	0.0415
Paint Application – Water Based	0.5138	0.1158	0.0428	0.3276
Chemical Manufacturing	0.727	0.1668	0.0052	0.101
Chemical Fertilizer – Urea	0.777	0.1759	0.0065	0.0406
Agricultural Tillage Dust	0.0205	0.0815	0.322	0.576
Livestock Dust	0.0399	0.0477	0.3671	0.5453
Feed and Grain Operations	0.0013	0.0283	0.2338	0.7366
Grain Drying	0.2598	0.1499	0.1167	0.4736
Coffee Roasting	0.4833	0.111	0.0202	0.3855
Cotton Ginning	0.4603	0.1047	0.0372	0.3978
Petroleum Refining	0.4382	0.1261	0.0377	0.398
Asphalt Roofing Manufacture	0.7782	0.1706	0.0301	0.0211
Asphaltic Concrete Batch Plant	0.2725	0.0725	0.044	0.611
Cement Prod./Concrete Batcing	0.2624	0.3622	0.2892	0.0862
Lime Manufacturing	0.0591	0.0658	0.153	0.7221
Calcination of Gypsum	0.164	0.3348	0.3722	0.129
Clay and Related Products Mfg.	0.4264	0.0979	0.0284	0.4473
Glass Melting Furnace	0.7893	0.1756	0.0143	0.0208
Fiberglass Forming Line	0.8059	0.1805	0.0074	0.0062

Table B.1, cont.

Profile Name	$PM_{< 0.625}$ Fraction	$PM_{0.625-2.5}$ Fraction	$PM_{2.5-10}$ Fraction	$PM_{> 10}$ Fraction
Steel Heat Treating – salt Quench	0.6323	0.2298	0.0953	0.0426
Steel Sinter Plant	0.793	0.1781	0.0085	0.0204
Steel Abrasive Blasting	0.653	0.1436	0.0582	0.1452
Steel Open Hearth Furnace	0.7668	0.1667	0.0454	0.0211
Basic Oxygen Furnace – Steel	0.8111	0.181	0.0078	0.0001
Electric Arc Furnace	0.3892	0.2168	0.2145	0.1795
Aluminum Foundry	0.7445	0.1632	0.0401	0.0522
Wood Operation – Sanding	0.7284	0.1619	0.0272	0.0825
Wood Operation – Resawing	0.1717	0.1235	0.0893	0.6155
Pulp and Paper Mills	0.01	0.1394	0.3267	0.5239
Mineral Process Loss	0.01	0.1394	0.3267	0.5239
Rock Crushers	0.0062	0.024	0.0549	0.9149
Rock Screening and Handling	0.01	0.1394	0.3267	0.5239
Landfill Dust	0.2427	0.1453	0.146	0.466
Road and Bldg. Construction Dust	0.2002	0.1767	0.246	0.3771
Paved Road Dust	0.0222	0.0586	0.3524	0.5668
Unpaved Road Dust	0.0367	0.0917	0.4623	0.4093
Tire Wear	0.162	0.1714	0.0548	0.6118
Windblown Dust – Agricultural	0.0206	0.0802	0.3733	0.5259
Windblown Dust – Unpaved Areas	0.0479	0.0738	0.3529	0.5254
Unspecified	0.048	0.074	0.3528	0.5252

Table B.2: Summary of PM_{10} composition distribution data (Kumar and Lurmann, 1996).

Profile Name	PM_{10} – SO_4 Fraction	PM_{10} – EC Fraction	PM_{10} – OM Fraction	PM_{10} – Other Fraction
Liquid Material Combustion	0.244	0.1465	0	0.5853
Fuel Combustion – Residual	0.4373	0.1727	0	0.2541
Fuel Combustion – Distillate	0.244	0.1465	0	0.5853
Utility Boilers-Residual	0.2879	0.1966	0	0.4846
Stat. I. C. Engine – Liquid Fuel	0.244	0.1465	0	0.5853
Stat. I. C. Engine – Gasoline	0.4472	0.1988	0	0.3479
Stat. I. C. Engine – Diesel	0.1439	0.0383	0	0.7766
Vehicular Sources – Gasoline	0.0228	0.134	0.4189	0.4181
Vehicular Sources – Diesel	0.0233	0.3155	0.5384	0.0817
Marine Vessels – Liquid Fuel	0.1439	0.0383	0	0.7766
Gaseous Material Combustion	0.2001	0.4999	0	0.3
Residential – Natural Gas	0.2001	0.4999	0	0.3
Stat. I. C. Engine – Gas	0.4472	0.1988	0	0.3479
Petroleum Heaters – Gas	0.4459	0.0662	0	0.4365
Solid Material Combustion	0.0263	0.2989	0	0.6711
Coal/Coke Combustion	0	0.0528	0	0.3242
Stat. I. C. Engine – Solid Fuel	0.0263	0.2989	0	0.6711
Wood Waste Combustion	0.0263	0.2989	0	0.6711
Other Waste Combustion	0.0263	0.2989	0	0.6711
Planned/Unplannd Forest Fires	0.0156	0.1011	0.4743	0.2863
Agricultural Burning	0.0156	0.1011	0.4743	0.2863
Unplanned Structural Fires	0.0003	0.2936	0	0.6847
Fireplaces	0.0094	0.2086	0.5659	0.1332
Aircraft – Jet Fuel	0.244	0.1465	0	0.5853
Orchard Heaters	0.244	0.1465	0	0.5853

Table B.2, cont.

Profile Name	PM$_{10}$ – SO$_4$ Fraction	PM$_{10}$ – EC Fraction	PM$_{10}$ – OM Fraction	PM$_{10}$ – Other Fraction
Incineration – Liquid Fuel	0.244	0.1465	0	0.5853
Incineration – Gaseous Fuel	0.2001	0.4999	0	0.3
Incineration – Solid Fuel	0	0	0	0.2849
Evaporation	0.0192	0.5272	0	0.4121
Coating Material Evaporation	0.0192	0.5272	0	0.4121
Paint Application – Oil Based	0.0192	0.5272	0	0.4121
Paint Application – Water Based	0.0134	0.3363	0	0.3227
Chemical Manufacturing	0.018	0	0	0.881
Chemical Fertilizer – Urea	0.0387	0.3165	0	0.6042
Agricultural Tillage Dust	0.0041	0.0049	0.0892	0.335
Livestock Dust	0.0037	0.0066	0.143	0.3014
Feed and Grain Operations	0.0013	0.0791	0	0.183
Grain Drying	0	0	0	0.5264
Coffee Roasting	0.0032	0.147	0	0.4643
Cotton Ginning	0	0	0	0.6022
Petroleum Refining	0.2612	0.0202	0	0.3206
Asphalt Roofing Manufacture	0.2214	0.2349	0	0.5226
Asphaltic Concrete Batch Plant	0.002	0.0424	0	0.3446
Cement Prod./Concrete Batcing	0.2185	0.1364	0	0.5589
Lime Manufacturing	0	0	0	0.2779
Calcination of Gypsum	0.4718	0.0182	0	0.381
Clay and Related Products Mfg.	0.003	0	0	0.5497
Glass Melting Furnace	0.5415	0.1138	0	0.3239
Fiberglass Forming Line	0.0054	0.2976	0	0.6908
Steel Heat Treating – salt Quench	0.238	0.0676	0	0.6518
Steel Sinter Plant	0.1935	0.1064	0	0.6797
Steel Abrasive Blasting	0.0003	0.0003	0	0.8542
Steel Open Hearth Furnace	0.388	0.1958	0	0.3951
Basic Oxygen Furnace – Steel	0.3999	0.2001	0	0.3999
Electric Arc Furnace	0.3282	0.1641	0	0.3282
Aluminum Foundry	0.1516	0.1232	0	0.673
Wood Operation – Sanding	0	0.3761	0	0.5414
Wood Operation – Resawing	0.002	0.1616	0	0.2209
Pulp and Paper Mills	0	0	0	0.4761
Mineral Process Loss	0.0025	0	0	0.4736
Rock Crushers	0.0004	0	0	0.0847
Rock Screening and Handling	0.0025	0	0	0.4736
Landfill Dust	0.0041	0.0049	0.0892	0.335
Road and Bldg. Construction Dust	0.0041	0.0049	0.0892	0.335
Paved Road Dust	0.0041	0.0049	0.0892	0.335
Unpaved Road Dust	0.0041	0.0049	0.0892	0.335
Tire Wear	0	0.3377	0	0.0505
Windblown Dust – Agricultural	0.0042	0.0021	0.0213	0.4465
Windblown Dust – Unpaved Areas	0.0041	0.0049	0.0892	0.335
Unspecified	0.0014	0.0002	0.021	0.452