International Institute for Applied Systems Analysis
Schlossplatz 1 • A-2361 Laxenburg • Austria
Telephone: (+43 2236) 807 342 • Fax: (+43 2236) 71313
E-mail: publications@iiasa.ac.at • Internet: www.iiasa.ac.at

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Airborne Fine Particulates in the Environment: A Review of Health Effect Studies, Monitoring Data and Emission Inventories

Matthias Koch

Approved by

Markus Amann
Project Leader
Transboundary Air Pollution Project

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Contents

Abstract

Airborne fine particles are a mixture of various components and are emitted by different sources. Short-term and long-term epidemiological studies have associated fine particles with adverse health effects, excess mortality, respiratory and cardiovascular diseases. There are indications that the health effects are more associated with the fine fraction of PM10 and with ultrafine particles (< 0.1 µm) than with the coarse fraction. Recently, diesel exhaust and vehicular emissions have been identified in epidemiological studies as important factors for explaining adverse health effects of fine particles; there are also indications for a biological mechanism. For acid aerosols and sulfate particles, a biological mechanism is proposed in the literature, but results from epidemiological studies are not entirely conclusive. Commonly, no threshold is found and a linear dose-response relationship is proposed.

The spatial and temporal distribution of fine particle levels may vary substantially. Low PM10 levels are found in remote areas at about 10 µg m^{-3}, heavily polluted urban areas may reach 100 µg m^{-3} on average. PM2.5 (< 2.5 µm in diameter) commonly comprises 60 % of PM10. Fine particles are emitted as primary pollutants and are also formed during secondary processes in the atmosphere, i.e., through the oxidation of SO_2, NO_x and VOC emissions. Fine particles are modeled deductively and inductively, i.e., via dispersion and receptor models, respectively. Emission inventories are still in an early stage and need careful consideration of adequate emission factors and other assumptions.

In urban areas, traffic is usually an important source of fine particles, although locally the situation can be dominated by emissions from local industries. For remote areas, where local sources are absent, regional and transboundary sources may be prevalent. We hypothesize that vehicular emissions and particularly diesel exhaust are likely to be important, if not the major, factors for the adverse health effects associated with fine particles.

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About the Author

Matthias Koch studied environmental science at Technische Universität Berlin (Germany), Edinburgh University (Scotland/UK) and Umeå Universitet (Sweden). He was a consultant with Prognos AG in Basel (Switzerland). At present, he is a research scientist at Technische Universität Berlin. His research interests focus on environmental pollutants, particularly source identification, flow analysis, integrated modeling and reduction strategies.

E-mail: matthias.koch@tu-berlin.de

Airborne Fine Particulates in the Environment: A Review of Health Effect Studies, Monitoring Data and Emission Inventories

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This work was carried out as part of IIASA's Young Scientists Summer Programme (YSSP) in 1998 and reflects the state of knowledge in 1998.

1. Introduction

It is well known for decades that air pollution causes adverse health effects. During smog periods, substantial amounts of excess deaths were reported, e.g., the London fog episodes (Thurston *et al,*. 1989). The association between high levels of ambient particles and human mortality has been well established in the past.

In the last decades, the emission of total suspended particles (TSP) and ambient levels of TSP have fallen substantially due to the implementation of air pollution control devices, i.e., filters, electric precipitators etc. Thereafter, it was expected that these measures would prevent further human health effects associated with particles. However, since the late 1980's epidemiological studies were published relating mortality and morbidity to ambient levels of fine particles. Most emphasis was then given to the particles with an aerodynamic diameter smaller than 10 µm (PM10).

In several countries (USA, the Netherlands, Sweden, UK), governmental commissions reviewed the evidence of the association between ambient levels of fine particles and human health effects (US-EPA 1996a, Gezondheitsraad 1995, Camner *et al*. 1992, COMEAP 1995). Even though the mechanism of action was not clear, the general conclusion was that ambient levels of airborne fine particles are associated with excess mortality and morbidity. Regulatory measures concerning ambient levels of fine particles were put forward in the US and in the European Union. The World Health Organization-Europe did not recommend specific thresholds in their guidelines, but suggested a continuum of effects with increasing exposure (WHO-Europe, 1995). Table 1 summarizes the air quality standards.

Governments commissioned some large-scale epidemiological studies on the link between air pollutants and human health: the Harvard Six Cities Study in the US and the APHEA and the PEACE projects in Europe. Emission inventories were established. The monitoring of fine particles in the environment was intensified.

Even though PM10 is only a fraction of TSP, it still consists of a mixture of many different constituents. PM10 is emitted by various sources. Established size fractions are: ultrafine particles (diameter < 0.1 µm), the fine fraction of PM10 with an aerodynamic diameter smaller than 2.5 µm (i.e., PM2.5) and the coarse fraction of PM10 with an aerodynamic diameter between 2.5 and 10 µm (PM10-2.5), see Figure 1. The sources of primary particles can be categorized as anthropogenic or biogenic, fugitive emissions.

Table 1 Air quality standards and guidelines for fine particles in various countries[1], in µg m^{-3}

Particle size	**Time period**	**USA**	**NL**	**Sweden**	**Brazil**	**Mexico**	**WHO**	**EU proposal**	
Reference		US-EPA 1996b	RIVM 1996	Natur-vardsverket 1997	Kretz-schmar 1994	Kretz-schmar 1994	WHO-Europe 1987		
Enforcement								until 2005	until 2010
PM10	Annual average	50	40	(20)	50			30	20
	Daily average	150	70	100	150	150	70	50	50
PM2.5	Annual average	15							
	Daily average	65							
Particulate strong acidity	Annual average								
	Daily average			10					

Fine particles are divided into primary and secondary particles. Primary particles are emitted by a source as particles and dispersed in the atmosphere without any major chemical transformation. Secondary particles are emitted as gaseous pollutant (e.g., SO_2, NO_x) and react chemically in the atmosphere to form particles. For instance, SO_2 is

[1] The detailed US standards are 15 µg m^{-3} for the 3-year-average of annual arithmetic mean PM2.5 concentrations from single or multiple community-oriented monitors, 65 µg m^{-3} for the 3-year average of the 98th percentile of 24-hour PM2.5 concentration at each population-oriented monitor within an area, 15 µg m^{-3} for the 99th percentile of 24-hour PM10 concentrations at each monitor within an area, 50 µg m^{-3} for the 3-year average of the annual arithmetic mean concentration.

The EU proposal for the PM10 24-hour average limit value is not to be exceeded more than 25 times per year until 2005 and more than 7 times per year until 2010.

The Swedish guideline of 20 µg m^{-3} is not an annual average but a half-year average for the winter period October until March.

oxidized to sulfate and forms with ammonia/ammonium ammonium sulfate particles. Fine particles are found to constitute of six major categories (US-EPA, 1996b): sulfates, nitrates, organic carbon, elemental carbon, trace metals, and water. Sulfates and nitrates are mainly secondary particles. For organic carbon, both primary and secondary formation is relevant. Elemental carbon and metals are usually emitted as primary particles. Table 2 gives an overview on the most relevant sources of primary and secondary fine particles.

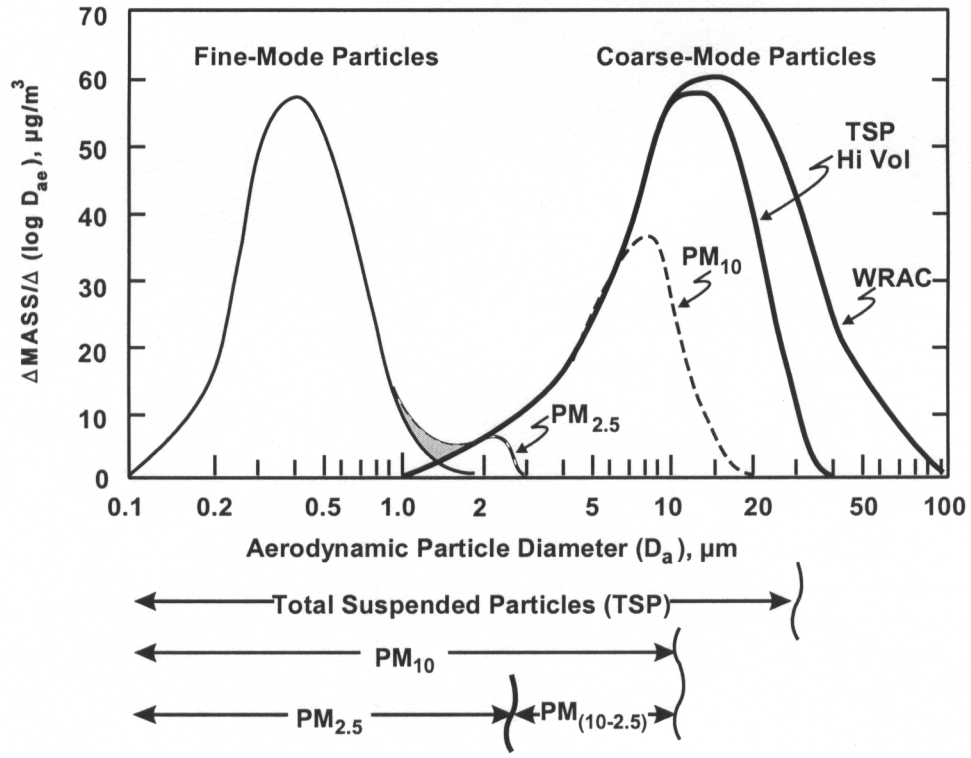

Figure 1 Size distribution of fine particles and fractions collected by size-selective samplers (US-EPA 1996b)

In general, for PM2.5 the secondary formation is more relevant than primary emissions. For PM10-2.5, the primary emissions should be considered in more detail than the secondary formation. PM10 is considered more a local and urban scale problem, whereas PM2.5 has a much longer residence time and is considered a regional or even a transboundary problem (see Figure 2). PM2.5 is highly correlated with PM10 but poorly correlated with PM10-2.5 (Wilson and Suh, 1997). The spatial variation is higher for PM10-2.5 than for PM2.5 (Burton *et al.* 1996).

Table 2 Sources of primary and secondary fine particles

	Transformation process	**Sources**
Primary particles	-	anthropogenic: fuel combustion, road transport, industrial processes, domestic heating, wood combustion natural: soil dust, sea salt
Secondary particles		
SO_4^{2-}	Oxidation of SO_2	Fossil fuel combustion, volcanic emissions
NO_3^-	Oxidation of NO_x	Fossil fuel combustion, vehicle exhaust
NH_4^+	Reduction of NH_3	Animals, sewage, motor vehicles
Elemental carbon	-	Motor vehicle exhaust, wood burning
Organic carbon	- / Oxidation of hydrocarbons	Chemical use, vegetation, motor vehicles

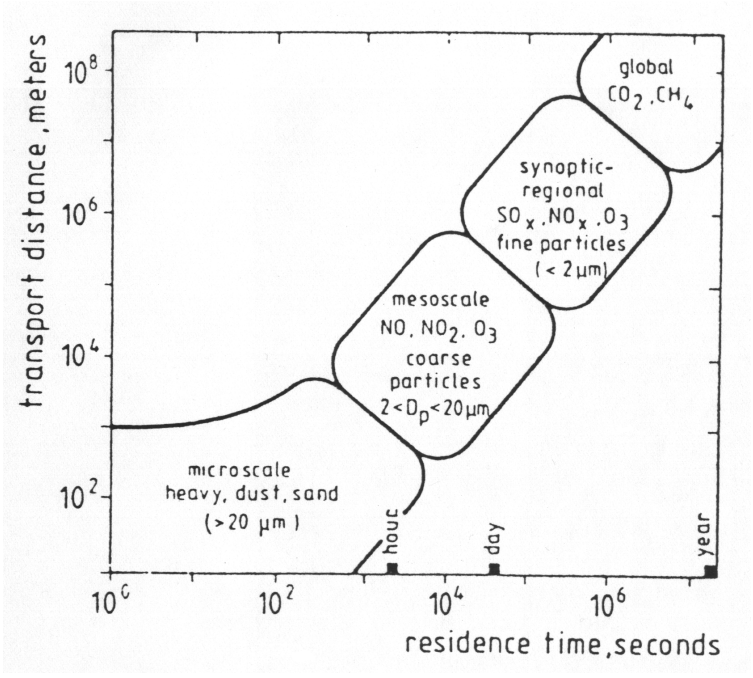

Figure 2 Travel distance and residence time of particles and gaseous pollutants (from: van Houdt 1990)

2. Health Effects

Health effects of fine particles are of concern in many countries, and a number of national review boards were established. Associations like the American Thoracic Society and the Air & Waste Management Association have set up communities or have commissioned reviews (N.N., 1996; Vedal, 1997). Several reviews are published in the scientific literature (Schwartz 1994a; Dockery and Pope 1994; Pope et al. 1995a; Ostro 1993).

Most of these commissions and reviews concluded that (i) there is an association between ambient levels of fine particles and human health effects, (ii) that this association is statistically significant, and (iii) that it can not be explained by confounding factors, even though the mechanism of action is not yet known and the specific constituent causing these effects has not yet been clearly identified.

Several dozens of epidemiological studies on the health effects of fine particles are published. In these studies, the population or subgroups, like the elderly, children or people with pre-existing illnesses, were investigated. The end points considered were mortality or morbidity, particularly respiratory and cardiovascular diseases. These were related to ambient levels of airborne fine particles, measured at one or several monitoring sites.

These studies were either conducted as short-term study, comparing time series of pollutant levels and health effects, or long-term studies, i.e., cross-sectional studies, comparing the effects and the factors between regions. Figure 3 gives an overview on the relevant aspects of these studies. Because usually only one aspect in each of the three groups was investigated, the results of these studies can not easily be compared.

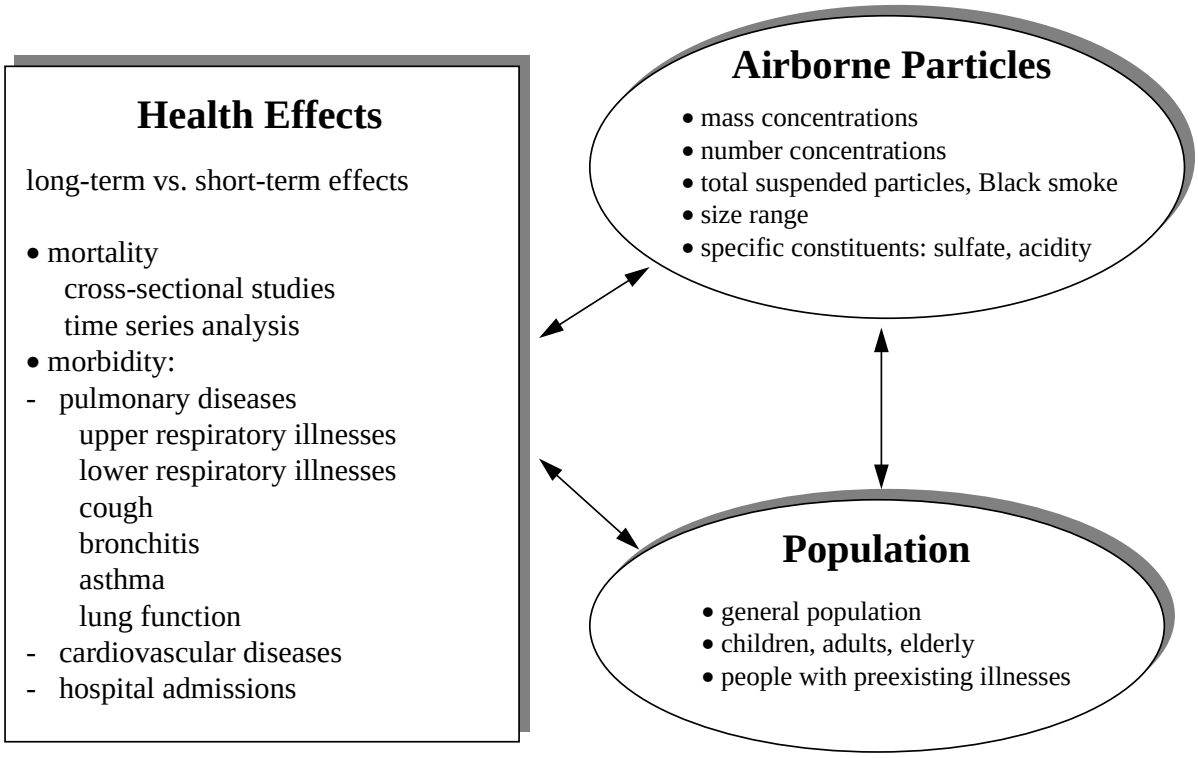

Figure 3 Structure and measurement scope of epidemiological studies

Numerous studies have been undertaken in North America, and some are available from Europe. The knowledge from Latin America and particularly from Asia is rather limited. Dockery and Pope (1994) reported a statistically significant 1 % increase (95 % confidence interval (CI): 0.7 %, 1.6 %) in daily mortality for each 10 µg m^{-3} increase in PM10 concentration in North America. No threshold was found. In the European APHEA project, the corresponding value was a 2 % increase (95 % CI: 1%, 3 %) in daily mortality for a 50 µg m^{-3} increase in PM10 concentrations (Katsouyanni *et al.,* 1997). The European PEACE project did not find a significant association between PM10 levels and the short-term changes in the health status of children with chronic respiratory symptoms (PEACE, 1998).

Several Latin American studies confirmed the associations of human health effects with ambient levels of airborne particulate matter (Ostro *et a,l.* 1996; Saldiva *et al,.* 1995; Borja-Aburto *et al.,* 1997; Romieu *et al.,* 1996; Romieu and Borja-Aburto, 1997), but not all studies (Saldiva *et al.,* 1994). In Asia, epidemiological research in this field is sparsely available. When extrapolating the American results, the excess mortality due to PM10 for the regions of Kathmandu Valley, Jakarta, Manila, and Mumbai was estimated 85, 4500, 1300, and 2765 cases, respectively (World Bank 1997a, b, c, d).

2.1 The Overall Evidence

The strength of the evidence, relating human health effects to airborne particles, has been discussed extensively in the scientific literature (Vedal 1997 and comments, Moolgavkar and Luebeck, 1996; Gamble and Lewis, 1996; Moolgavkar *et al*. 1995a; Dockery and Schwartz, 1995; Moolgavkar *et al*,. 1996; Moolgavkar *et al.* 1995b; Lipfert and Wyzga, 1995; Samet *et al.* 1998; Moolgavkar 1994; Kaiser 1998). The major criticism addressed the question of inadequate adjustment for confounding factors, i.e., weather and climate, co-pollutants and age. It was also criticized that the monitoring data inadequately represents the population exposure, that the biological mechanism of action is still not known and that earlier exposures could disturb the investigation, i.e., if the population had already been exposed to particles before the epidemiological study started.

The value of epidemiological studies can be limited because of deficiencies in controlling the effect of confounding co-pollutants, that show either a high correlation or that have not been measured at all (Moolgavkar and Luebeck, 1996). In time-series studies, it is not an easy task to distinguish the effect of one pollutant from the effect of other pollutants, when they are highly correlated. This can be the case for other air pollutants, i.e., SO_2, NO_x, ozone, CO. Particularly ultrafine particles and PM2.5 are usually well correlated with SO_2. In cross-sectional studies, this is less problematic when comparing regions with very different pollutant levels. In several US communities with low sulfur dioxide and ozone concentrations e.g., Utah Valley (Pope, 1996), the association between fine particle pollution and human health was still significant and could not be explained by confounding co-pollutants (Dockery and Schwartz, 1996).

The effect of temperature, weather and climate on mortality has been known for a long time (Ellis, 1972). The epidemiological studies on the association between airborne fine particles and mortality have been criticized because of inadequately addressing the effect of weather on mortality (Gamble and Lewis, 1996; Lipfert and Wyzga, 1995; Moolgavkar and Luebeck, 1996; Pope and Kalkstein, 1996). Samet *et al.* (1998) showed that applying alternative approaches to control for temperature and weather had little effect on the association between mortality and air pollution. It was also shown that the association was independent of the region for different climate and weather characteristics (Schwartz 1993).

Two major factors can disturb the extrapolation from central monitoring data to personal exposure. The spatial distribution of the pollutant within one study area may not be represented appropriately by relying on central monitoring data. This is of particular concern for the coarse fraction of PM10, where spatial variability is known to be higher than for the fine fraction (Lipfert and Wyzga, 1997). Because human beings spend most of the time indoors rather than outdoors, indoor concentrations are much more relevant than outdoor concentrations. The indoor-outdoor relationship and the infiltration rates were discussed in detail by Wilson and Suh (1997). All these factors will increase the error when assuming that central monitoring data represent population exposure adequately. This is particularly problematic for the coarse fraction of PM10.

Emphasizing the missing or inadequately addressed confounding factors is in the sense convincing that noise in the epidemiological data and uncertainties still need to be kept in mind, but it is not convincing in the sense that the observed effects can only be explained by confounding factors. Finally, it remains a political question how strong the evidence needs to be until action is taken and how much uncertainty is still accepted.

2.2 Size Ranges

Because PM10 and total suspended particles (TSP) consist of a broad mixture of primary and secondary particles from various sources with different kinds of properties, the identification of the specific size range and constituents most likely to be associated with the observed health effects was and still is a critical task.

When investigating the role of PM2.5, PM10-2.5, sulfate and acidic particles within the Harvard six cities study, Schwartz *et al.* (1996) found that the daily mortality was much more associated with the fine fraction (PM2.5) than with the coarse fraction (PM10-2.5). A similar weaker association was also found for sulfate particles, but not for H^+ concentrations. Lipfert and Wyzga (1997) and White (1998) criticized these findings of being biased by differential measurement errors, i.e., that the measurement error for the coarse fraction is substantially higher than for the fine fraction and that this would lead to misattribute effects of coarse particles to fine particles. But this effect was only discussed qualitatively without quantifying the effects.

The effect of fine and ultrafine particles on the respiratory system of asthmatics was investigated in a German study by Peters *et al.* (1997) and in a Finnish study by Pekkanen *et al.* (1997). A size range between 0.01 µm to 10 µm was considered. Peters *et al.* (1997) showed that a decrease in morning peak expiratory flow was best associated with the concentration of ultrafine particles (0.01 – 0.1 µm), particularly with their number concentration. Pekkanen *et al.* (1997) observed a statistically significant association for PM10 and black smoke, but not for the smaller size fractions. It was suggested that this difference might be explained by the difference in their sources. In the German city of Erfurt, combustion of brown coal was the major particle source, whereas in Kuopio (Finland) traffic exhausts were dominating.

There is some indication that human health effects are more associated with the fine fraction than with the coarse fraction and that specifically the ultrafine particles - as hypothesized by Seaton *et al.* (1995) - are of major concern, but some uncertainty on the size range mainly responsible remains.

2.3 Constituents

Fine particles do not only vary in size but also in their constituents, chemical composition, physical properties etc. There are less epidemiological studies available having measured particle constituents compared to the number of studies without information on particle constituents. Sulfate and acidity are covered in some studies, other constituents like metals and diesel exhaust or a comparison of crustal vs. combustion related particles have rarely been studied.

The acidity of fine particles was the constituent most frequently investigated (US-EPA 1996b; Vedal 1997). Investigations of historical air pollution episodes showed an association between acid aerosol concentrations and mortality (Thurston *et al.,* 1989). The association between particulate acidity and human health effects was shown for several types: the peak expiratory flow of children (Raizenne *et al.,* 1996), respiratory hospital admissions (Thurston *et al*., 1994), emergency room visits of children (Delfino *et al.,* 1997). Other studies did not find a significant association for the respiratory symptoms of children (Schwartz *et al.,* 1994), and mortality (Dockery *et al.,* 1992, Schwartz *et al.,* 1996). The US-EPA (1996b) concluded that chronic exposure might have effects on the respiratory system of children. The overall picture is not very clear, particularly whether or not acidity plays an important role in the health effects of fine particles. Because of the high correlation to PM2.5 in time series studies, it is particularly difficult to clearly distinguish these from each other.

Sulfate has shown positive and statistically significant associations with the respiratory health (Lippmann and Thurston, 1996). Several studies in Canada and the US have shown the effect of sulfate on human health: on mortality (Schwartz *et al,.* 1996, Pope *et al.*. 1995b, Dockery *et al,.* 1993), on hospital admissions (Burnett *et al.,* 1995, *Burnett et al.,* 1994), on respiratory health of children (Dockery *et al*., 1996, Dockery *et al.,* 1989) and on emergency room visits (Delfino *et al*., 1997). A Dutch study did not show an association with acute respiratory symptoms in children (Hoek and Brunekreef, 1995). Although an association between sulfate and respiratory illnesses seems well documented, a related cause-effect relationship may be flawed by the strong correlation with PM2.5 and acidity. A biological mechanism was proposed for acidity, but not yet for PM2.5.

Diesel exhaust may cause human illnesses, including cancer (Cox 1997; Bhatia *et al,* 1998; Pott and Roller, 1997). Recent investigations in the Netherlands found an association between living distance from motorways or other streets with high traffic density and respiratory symptoms in children (Oosterlee *et al.*, 1996; Wjst *et al.,* 1993; Duhme *et al.,* 1996). A British study did not show an association between proximity to a motorway and asthma in children (Waldron *et al.,* 1995). At a living distance less than 100 m (van Vliet *et al.,* 1997) and 300m (Brunekreef *et al.,* 1997) from a motorway, significantly more chronic respiratory illnesses in children were reported. The lorry traffic density, but not the passenger traffic density, on a motorway was significantly associated with respiratory symptoms in children (de Hartog *et al.,* 1997; *Brunekreef et al.,* 1997).

Diesel exhaust is a most convincing constituent of fine particles associated with adverse health effects. Toxicological tests also showed respiratory diseases caused by diesel exhaust, even though these were not necessarily identical to those effects found in the epidemiological studies. The majority of epidemiological studies also supports that sulfate particles are associated with adverse health effects, but no biological mechanism is yet available and a high correlation to fine or ultrafine particles may be responsible instead. Particulate strong acidity has been shown to affect human health at high levels in the past, but whether this holds true for lower concentration levels observed now is not clear.

2.4 Biological Mechanism

When investigating cause effect relations of exposure-related health issues, epidemiological studies are a very useful tool for identifying possible associations. However, for obtaining evidence also toxicological studies should be taken into account. The accurate simulation of ambient fine particles in toxicological studies is a challenge because they are a mixture of different components with broad toxicological characteristics and composed of various sources. The goal of the toxicological studies would be to identify specific components or sources that are mainly responsible for the health effects. Ultrafine particles, acidic sulfates as well as diesel exhaust and vehicular emission have been brought forward (Schlesinger, 1995). Oberdörster *et al.* (1995) found that ultrafine particles below a diameter of 0.1 µm caused increased pulmonary inflammatory responses in animal studies. Acidic sulfates have been shown in animal studies to induce nonspecific airway hyperresponsiveness (el-Fawal and Schlesinger, 1994) and other pulmonary effects (Amdur and Chen, 1989). Diesel exhaust and other vehicular emissions cause lung function deficiencies in human and animals (Mauderly, 1994; Scheepers and Bos, 1992a and 1992b). Fine particles from vehicular sources had a higher mutagenic effect than samples from other sources (Bronzetti *et al.,* 1997; Stevens *et al.,* 1990). When comparing these three components, it remains unclear which of these is closest to the effects shown in epidemiological studies. The specificity of the effects found in epidemiological studies was best seen for acidic sulfate. For diesel exhaust and vehicular emissions, toxicological effects were also shown for „real world“ samples.

2.5 Thresholds, Dose Response Relationship and Health Costs

For the development of abatement strategies it is important to understand dose-response relationships and, if possible, define appropriate critical levels below which effects do not occur according to present knowledge. Several studies found health effects even at low ambient levels of particles (Brandli, 1996; Hoek *et al.,* 1997; US-EPA 1996b). No threshold at all or, if any, below common ambient levels was concluded in a number of studies (Schwartz, 1993; Schwartz, 1994b; Wordley *et al.,* 1997; Brandli, 1996; Hoek *et*

al., 1997). The dose-response relationship was commonly found to be linear (Pope and Kalkstein, 1996; Schwartz 1994b; Wordley *et al.*, 1997; Hoek *et al.*, 1997). The WHO-Europe did not recommend a guideline value but suggested instead an exposure-response approach (WHO-Europe, 1995).

The health effects associated with fine particles also affect the society and the economy. Adverse productivity effects on labor force, particularly sickness leaves, have been found for Norway (Hansen and Selte, 1997; Rosendahl, 1998). Excess mortality and morbidity results in higher costs for health services. Pearce and Crowards (1996) estimated for the UK 7000 deaths annually and £14 billion annual health costs attributed to anthropogenic PM10. In the United States, the enforcement of the new PM2.5 standard is estimated to bring health benefits in the order of $32 billion annually (Ostro and Chestnut, 1998). Globally, 8 million additional deaths between 2000 and 2020 were estimated when comparing a business-as-usual and a climate change control policy (N.N. 1997). The link between climate change policy and excess mortality was obtained through the emission reductions of particulate matter and their health effects.

Overall, the emission control of fine particles does not only impose additional costs to industry and other emission generating economic sectors but will also have positive economic effects in terms of health benefits and reduced sickness leaves for the economy and society as a whole.

3. Monitoring of Particulate Concentrations in the Atmosphere

Present efforts for monitoring of fine particles is not evenly distributed throughout the world. TSP measurements are readily available in most countries. By using relative ratios, i.e. the proportion of specific size ranges relative to TSP, levels of PM10 and PM2.5 have been estimated. Instead of applying this approach, we reviewed the literature about direct measurements of PM10, PM2.5 and their constituents throughout the world.

Until the late 1980's, most of PM monitoring was done in North America; while much less is available from other continents. Because the analytical techniques were not very well developed until then, these studies are not outlined in detail here. Instead, the studies published since the beginning of the 90's are discussed in detail, focussing on Europe, Latin America and Asia, where the amount of research in this field is still limited. Finally, a general overview about the observed levels of fine particles in the environment, the temporal variation, their components, the contributing sources and the modeling approaches is given.

3.1 Europe

Until recently, continuous monitoring of fine particles is established only in a few European countries. In 1999, however, the EU Air Quality Daughter Directive on SO_2, NO2, particulate matter and lead (99/30/EC) obliged countries to regularly monitor PM levels and to report to the European Commission.

In the present data, PM10 measurements are much more common than measurements of PM2.5 or specific constituents. The UK is monitoring PM10 at 16 sites since 1995 (QUARG, 1996) and at some more sites since 1998. PM2.5 is measured at four sites since the beginning of 1998. In Norway, Sweden and the Netherlands PM10 was monitored for some years at several sites, particularly in urban areas. In Germany, the state Baden-Württemberg is monitoring PM10 at four sites and PM2.5 at one site since 1996/97 (Siegel, 1998).

When the PEACE study was undertaken, PM10 levels were measured at twelve urban and control locations in various countries in Europe during the winter of 1993-94. The observed average concentrations during this period were between 11 and 99 $\mu g\ m^{-3}$ (see Table 3). Lowest values were observed in Scandinavia, with PM10 concentrations between 10 and 20 $\mu g\ m^{-3}$. In Germany and the Netherlands concentrations between 33 and 52 $\mu g\ m^{-3}$ were measured. In Central Europe (i.e., Czech Republic, Poland and Hungary) up to 74 $\mu g\ m^{-3}$ were measured. In southern Europe, the observed average concentrations were between 50 and 99 $\mu g\ m^{-3}$.

Table 3: European PM10 levels in winter 1993-94, in µg m^{-3} (PEACE 1998)

	Urban location	**Control location**
Umea, Sweden	13.4	11.5
Southern Sweden	22.9	16.2
Kuopio, Finland	17.7	13.0
Oslo, Norway	19.3	11.2
Amsterdam, The Netherlands	45.3	44.4
Berlin, Germany	52.3	43.0
Eastern Germany	40.3	32.9
Teplice, Czech Republic	74	32
Prague, Czech Republic	52.7	49.6
Katowice, Poland	68.7	73.8
Krakow, Poland	60.1	56.1
Hungary	60.9	52.1
Pisa, Italy	61.8	69.5
Athens, Greece	98.8	50.0

The monitoring data from selected European studies are summarized in Table 4.

Table 4 Concentrations of fine particles in Europe, in $\mu g\ m^{-3}$

Location	**Rome, Italy**	**Norwich, UK**	**Ljublana, Slovenia**	**Erfurt, Germany**	**Birming-ham UK**	**Switzer-land**	**Wage-ningen, Nether-lands**	**Arnhem, Nether-lands**	**Erfurt, Ger-many**	**Sokolov, former CSSR**	**Nether-lands**
Source	D'Innocenzio 1998	Nicholson and Davies 1990	Bizjak *et al.* 1988	Tuch *et al.* 1997	Harrison *et al.* 1997	Monn *et al.* 1995	Janssen *et al.* 1997		Cyrys *et al.* 1995		RIVM 1996
TSP	52										
PM10	37				20	10 (alpine) 33 (urban)	39 (street) 32 (back-ground)	75 (street) 62 (back-ground)	60	54	35 (Range of mean con-centrations from 10 monitoring sites)
PM10-2.5	16										
PM2.5	22			74	16		43 (street) 35 (back-ground)				
H^+									0.3	0.4	
SO_4^{2-}		8 - 12	30 – 80						9.8	8.9	
Details	urban air, 44 samples, 1 sampling site		winter period								

In 1993-94, the annual average PM10 concentrations at the continuous monitoring sites in the UK were usually 20 – 34 µg m^{-3}, with slightly higher concentrations at sites with large traffic densities and domestic coal burning. Maximum hourly concentrations reached several hundred µg m^{-3}.

Harrison *et al.* (1997) monitored PM10 and PM2.5 in Birmingham (UK) and investigated major sources between October 1994 and October 1995. The mean PM10 concentrations were approximately 20 µg m^{-3} at three sites in the city center, at a background site and close to a motorway; i.e. the PM10 concentrations were essentially similar at all three sites. The mean PM2.5 concentration at the site close to the motorway was 16 µg m^{-3}. PM10 mainly consisted of three distinct components: Primary vehicular emissions, secondary pollutants and wind blown dust. Vehicle exhaust contributed 32 % of PM10, 41 % of PM2.5 and negligible to the coarse fraction. The fine fraction was primarily derived from vehicle emissions and secondary aerosols. The coarse fraction was mainly composed of re-suspended dust. In winter, particulate pollution episodes were primarily related with elevated levels of the fine fraction from vehicular emissions, whereas summer episodes were more related to wind-blown dust affecting the coarse fraction.

Nicholson and Davies (1990) analyzed sulfate particles and SO_2 sampled in Norwich (UK) over a period of nine months. Mean sulfate concentrations were 8 – 12 µg m^{-3} for the five sites. The results indicate that a distinct proportion of the SO_4^{2-} has been transported via long distances from the east.

In Sweden, PM10 was measured by the Swedish Environmental Protection Agency and by Stockholm University. The observed half-year averages were about 15 µg m^{-3} with maximum daily averages of 30 – 60 µg m^{-3} from 1993 until 1997 (Naturvardsverket, 1997). The diurnal profile shows that during summer the highest concentrations occur around noon, the minimum concentrations are reached in the night around 3 a.m. During winter, the diurnal variation is less pronounced (Areskoug *et al.,* 1995).

Annema *et al.* (1996) reviewed the research and monitoring of fine particles in the Netherlands during 1994. The overall mean PM10 concentration was 35 µg m^{-3}, ranging between 30 and 42 µg m^{-3} at each of the 10 monitoring stations between February 1992 and July 1993. During winter daily mean concentrations reached more than 100 µg m^{-3} , but very rarely in summer. No substantial diurnal cycle was found. The one hour concentrations were considerably higher at low wind speed compared to higher wind speed. The fine particles consisted of primary particles (25 – 30 %), secondary nitrate (25 – 30 %), secondary sulfate (15 – 20 %) and ammonium (15 – 20 %). Neighboring countries contribute substantially to the ambient levels in the Netherlands: Germany (35 %), Belgium, France, and the UK (10 % each). The Netherlands itself contributed only 20 %.

Janssen *et al.* (1997) investigated the effect of traffic on the levels of PM10 and PM2.5 by comparing the levels near a busy road and a background location in the Netherlands during 1994. In Arnhem, PM10 average concentrations of 75 µg m^{-3} and 62 µg m^{-3} were measured at street sites and background sites, respectively. The corresponding PM2.5 average concentrations were 43 µg m^{-3} and 35 µg m^{-3}, respectively. In Wageningen, PM10 average concentrations were 39 µg m^{-3} and 32 µg m^{-3} for the street and

background samples, respectively. The levels in the street samples were generally 30 % higher than in the background samples. The greater part of this difference for the PM2.5 could be related to elemental carbon. This emphasizes the role of diesel exhaust emissions.

Kainka *et al.* (1997) measured PM10 and PM2.5 at four locations in Germany. Single particle analysis revealed that surrounding sources were reflected in the coarse fraction whereas soot particles and soot agglomerates were more or less present in all locations.

Brand *et al.* (1991) measured the particle mass and number concentrations of the size range 0.01 – 25 µm at a remote site and at an urban site in Germany. The particle number concentration was about 5000 cm^{-3} and 32000 cm^{-3} at the remote site and at the urban site, respectively; the corresponding mass concentrations were 38 µg m^{-3} and 71 µg m^{-3}, respectively. The particle number concentration was higher at the urban site below a particle diameter of 2 µm and higher at the remote site above 2 µm.

Tuch *et al.* (1997) measured particle mass and number concentrations of the size range 0.01 – 2.5 µm for a period of 180 days during the winter 1991/92 at a site in eastern Germany, which is directly influenced by brown coal combustion. The particle number concentration was 18000 cm^{-3}. It was dominated by the fraction smaller than 0.1 µm (72%). The size range of 0.1 - 0.5 µm governed the particle mass concentration (83%), the mass concentration was 74 µg m^{-3}. The mass and number concentrations were poorly correlated.

Cyrys *et al.* (1995) measured sulfate, acidity, and PM10 concentrations in Erfurt (Germany) and Sokolov (Czech Republic) from December 1990 until June 1992. The mean PM10 concentrations were 60 µg m^{-3} and 54 µg m^{-3}, the mean SO_4^{2-} concentrations 9.8 µg m^{-3} and 8.9 µg m^{-3} and the H^+ concentrations 0.3 µg m^{-3} and 0.4 µg m^{-3} for Erfurt and Sokolov, respectively.

Monn *et al.* (1995) measured PM10 and TSP at for a period of one year in 1993 urban, rural, and alpine sites in Switzerland. The PM10 concentrations ranged between 10 µg m^{-3} at alpine sites and 33 µg m^{-3} at urban sites. The PM10 concentration showed a good correlation with NO_2 and SO_2 at urban sites, indicating similar emission sources.

D'Innocenzio *et al.* (1998) measured urban air at one monitoring site in Italy for the period October 1995 until September 1996. The concentrations of PM2.5 and PM10 were substantially higher during the winter period compared to the summer period, while only little difference was seen for PM10-2.5 and TSP. The ratio of PM10-2.5/PM2.5 was 0.97 and 0.68 in the winter period and in the summer period, respectively.

Bizjak *et al.* (1988) investigated the sources of sulfate particles in Ljubljana (Slovenia, former Yugoslavia). Three distinct periods were found within the diurnal cycle: (i) heterogeneous, aqueous oxidation of local SO_2 was attributed to the sulfate particles in the morning; (ii) in the evening, sulfate particles were from primary emission; (iii) during the other periods, heterogeneous and homogeneous oxidation of regional SO_2 was responsible.

In Europe, annual average PM10 concentrations were usually around 30 µg m^{-3} and averages in the winter period at about 50 µg m^{-3}. In Scandinavia and in remote sites in

Switzerland, concentrations between 10 and 20 µg m^{-3} were measured. Very high concentrations (up to 100 µg m^{-3}) were typically found in eastern and southern Europe.

Few measurements of PM2.5 have been reported in Europe: 11 – 16 µg m^{-3} as monthly averages in Birmingham (UK), about 40 µg m^{-3} in Arnhem (The Netherlands), 22 µg m^{-3} in Rome (Italy) and 74 µg m^{-3} in Erfurt (Germany). Sulfate concentrations were typically about 10 µg m^{-3}. Particle number concentrations were only reported from Germany: 5000 cm^{-3} at a remote site in the North Sea, 32000 cm^{-3} in Frankfurt/Main.

3.2 Latin America

No data from continuous monitoring stations in Latin America were found. We therefore only report single studies from Brazil, Chile, Mexico, and Puerto Rico. Table 5 gives an overview on fine particle monitoring data in Latin America.

Table 5 Concentrations of fine particles in Latin America, in µg m^{-3}

Location	**Mexico City**	**Santiago de Chile**	**Mexico City**	**Sao Paulo**	**Sao Paulo**
Source	Salazar *et al.*, 1992	Rojas *et al.*, 1990	Vega *et al.*, 1997	Andrade *et al.*, 1993	Kretzschmar *et al.*, 1994
Year			1989-90	1989	
TSP			210		
PM10		100			70-150 (1981) 20-100 (1992)
PM10-2.5	36 / 29	66	96	61	
PM2.5	33 (dry season)23 (wet season)	34		37.5	

Kretzschmar *et al.*, (1994) reported PM10 levels in several Latin American cities. Average PM10 concentrations of 16 sites in Sao Paulo were between 70 and 150 µg m^{-3} in 1981 and 20 – 100 µg m^{-3} in 1992. The general trend was decreasing; while at some sites there was a dramatic reduction, at other sites there was hardly any change. In Rio de Janeiro, PM10 levels averaged 70 µg m^{-3} during the measurement period in winter 1984.

Salazar *et al.*, (1992) measured the particle concentrations for five size cuts in Mexico City. For the fine fraction (0.49 – 3 µm), 33 µg m^{-3} and 23 µg m^{-3} were measured as geometric mean in the dry and rainy season, respectively. The corresponding concentrations for the coarse fraction (3 µm - > 7.2 µm) were 36 µg m^{-3} and 29 µg m^{-3}. The concentration was lowest for the size range 1.5 – 3 µm (about 10 µg m^{-3}), much higher for the coarser fractions 3 – 7.2 µm and > 7.2 µm and moderately higher for the finer fractions 0.49 – 0.95 µm and 0.95 – 1.5 µm.

For the winter 1989/1990, Vega *et al.* (1997) investigated the sources of fine particles (PM2.5) in Mexico City. A PM2.5 average concentration of 96 µg m^{-3} was measured. The TSP average concentration was 210 µg m^{-3}. Major contributors to PM2.5 were gasoline vehicles without catalytic converter at approximately 40 %, followed by oil refineries (about 15 %) and re-suspended dust (15 – 20 %). Secondary aerosols (sulfate, nitrate, and organic carbon) contributed about 10 %, 2 – 5 %, and 3 – 5 %, respectively. Heavy-duty diesel vehicles were responsible for approximately 5 %. Because the samples were taken before regulatory action (catalytic converters, closure of a refinery etc.) were put in place, they might not reflect the current situation.

Andrade *et al.* (1993) investigated the sources of fine and coarse mode particles (PM2.5, PM15-2.5) for Sao Paulo, Brazil, by means of receptor modeling. In 1989, mean concentrations of PM2.5 and PM15-2.5 were 37.5 µg m^{-3} and 61 µg m^{-3}, respectively. Combustion of residual oil and diesel, soil dust, and industrial emissions accounted for 41 %, 28 % and 13 % of PM2.5, respectively. For the coarse particles, soil dust, industrial emissions, sea salt, and oil burning were contributing 59 %, 19 %, 14 %, and 8 %, respectively.

Rojas *et al.* (1990) investigated the sources of fine and coarse particles in Santiago de Chile. For the fine fraction (< 2.5 µm) and for the coarse fraction (2.5 – 15 µm), 34 µg m^{-3} and 66 µg m^{-3}, respectively, were measured. Secondary sulfate particles were responsible for approximately 50 % of the fine fraction, wood burning/car exhausts and residual oil combustion contributed 25 % and 13 %, respectively.

Infante *et al.* (1990) investigated the particles size distribution in Ponce (Puerto Rico) in 1985 – 1987 for the size ranges > 7 µm, 3.3 – 7 µm, 2 – 3.3 µm, 1.1 – 2 µm, and < 1.1 µm. The corresponding arithmetic mean concentrations were 24 µg m^{-3}, 12 µg m^{-3}, 5 µg m^{-3}, 4 µg m^{-3} and 8 µg m^{-3}, respectively. The TSP concentration was dominated by the size fraction > 7 µm, over 75 % of TSP is of the size fraction > 3.3 µm.

Only very few studies on airborne fine particles are available in Latin America. The urban concentrations in Latin America are similar and higher compared to those at urban sites in southern Europe. No measurements from rural sites were available. PM2.5 concentrations were about 30 µg m^{-3} in Mexico City and 61 µg m^{-3} in Sao Paulo.

3.3 Asia and Oceania

For Asia, no monitoring data on fine particles from continuous measurements were found. Instead studies from Japan, Taiwan, Indonesia and Saudi Arabia are outlined in the following (see Table 6). A study from Australia is also included here.

Chan *et al.* (1997) investigated levels and sources of airborne particulate matter PM2.5 and PM10 in Brisbane, Australia. The average PM10 concentration of five sites was 27 µg m^{-3}. For one site PM2.5 and PM10-2.5 in Brisbane were also measured: 7.3 µg m^{-3} and 10.4 µg m^{-3}, respectively. In other Australian cities, PM2.5 concentrations of 8 µg m^{-3} and 17 µg m^{-3} were found in New South Wales and Footscray (Melbourne), respectively. By using the elemental composition as a source indicator, crustal matter (from local industries, soil etc.), organic matter, sea salt, elemental carbon and ammonium sulfate contributed 25 %, 17 %, 12 %, 10 %, 7 %, to the PM10 concentrations, respectively. The site with high traffic load had a higher content of elemental carbon (17 %) and less crustal matter (18%). In the single PM2.5 sample elemental carbon, organics, Pb/Br and sulfate components were enriched relative to the PM10 samples, whereas sea salt and crustal components were less abundant. Some influence from local sources (traffic, industry) was found, but in general anthropogenic sources were relatively evenly distributed.

Table 6 Fine particle concentrations in Asia and Oceania, in µg m^{-3}

Location	**Taipei, Taiwan**	**Riyadh, Saudi Arabia**	**Brisbane, Australia**	**New South Wales, Australia**	**Mel-bourne**	**Sapporo, Japan**	**Taiwan**
Source	Li *et al.*, 1993	El-Shobokshy *et al.*, 1990	Chan *et al.*, 1997			Kaneyasu *et al.*, 1995	Chen *et al.*, 1997
TSP	70- 270						
PM10 or PM15	150	639	27 (five sites)			25	
PM10-2.5 or PM15-2.5		275	10.4 (one site)				30 – 47
PM2.5	86	370	7.3 (one site)	8	17		15 – 48

Kaneyasu *et al.* (1995) investigated the composition of PM10 and its seasonal variation in Sapporo, Japan during an 18-month period in 1987-1988. The PM10 concentrations were about 25 µg m^{-3} at two sites. Organic carbon, from direct emissions and from secondary formation, contributed about 17 % to PM10, and elemental carbon, mainly from automobile exhaust and domestic heating, about 14 %. SO_4^{2-} was about 16 % of PM10, NO_3^- were responsible for 4 % of PM10. A seasonal maximum in April/June

indicated the role of tropospheric ozone, i.e., as a nighttime chemical processes. Cl^-, mainly from sea salt, contributed about 2 %.

When investigating the influence of particles of outdoor air on indoor air quality in Taipei, Taiwan, Li *et al.* (1993) measured TSP, PM10 and PM2.5 concentrations in the vicinity of three residences. The average concentrations (n=29) were 150 µg m^{-3} and 86 µg m^{-3} for PM10 and PM2.5, respectively. The indoor concentrations were generally lower than the outdoor concentrations, indicating an infiltration of outdoor air.

Chen *et al.* (1997) investigated the source contribution to PM2.5 and PM10-2.5 at three sites in Taiwan in the period 1992 – January 1994. Mean concentrations for 2-6 day periods were 30 – 47 µg m^{-3} and 15 – 48 µg m^{-3} for the coarse and the fine fraction, respectively. The main sources for the coarse fraction were soil dust and (paved) road dust (21 – 42 %), burning of agricultural wastes (31 – 40 %), coal and fuel oil combustion (7 – 27 %), and marine aerosols (3 – 11 %). The main sources for the fine fraction were burning of agricultural wastes (31 – 39 %), sulfates (16 – 26 %), coal and oil combustion (11 – 19 %), and diesel exhaust (7.5 – 12 %).

Zou and Hooper (1997) investigated the particle size distribution and the morphology of airborne particles in Jakarta, Indonesia. The size ranges of 0.02 – 0.95 µm, 0.95 - 1.5 µm, 1.5 – 3 µm, 3.0 – 7.2 µm, > 7.2 µm were associated with 48 %, 9 %, 9 %, 17 %, and 17 % of TSP, respectively. The particles were bimodal lognormal distributed, maxima were at 0.6 µm and 6.3 µm. Zn, Pb, and K were more abundant in the fine fraction. Ca, Si, Al, and Mn were relatively evenly distributed and Na was more abundant in the coarse fraction than the other elements.

Sharma and Patil (1992) investigated the size distribution of airborne particles in Bombay, India. For the size ranges < 0.1 µm, 0.1 – 0.2 µm, 0.2 – 0.4 µm, 0.4 - 0.8 µm, 0.8 – 1.6 µm, 1.6 – 3.2 µm, 3.2 – 6.4 µm, 6.4 – 12.5 µm, 12.5 – 25 µm, > 25 µm the following mass concentrations were measured: 7 µg m^{-3}, 13 – 18 µg m^{-3}, 4 – 8 µg m^{-3}, 7 – 11 µg m^{-3}, 8 – 13 µg m^{-3}, 11.5 µg m^{-3}, 16 – 21 µg m^{-3}, 6 – 13 µg m^{-3}, 3 – 4 µg m^{-3}, 7 – 8 µg m^{-3}, respectively. The mass size distribution was bimodal, but tended towards trimodal for the industrial site. Mass distribution maxima were at 0.1 µm and 2 µm for the background site and at 0.05 µm, 0.3 µm, and 2 µm for the industrial site.

El-Shokobokshy *et al.* (1990) measured the fine and coarse fraction (PM2.5 and PM15-2.5) in Riyadh, Saudi Arabia. The mean concentrations in March - May 1988 were 379 µg m^{-3} and 275 µg m^{-3} for PM2.5 and PM15-2.5, respectively.

Because the few available measurements from Asia, the conclusions that can be drawn are limited. The highest concentrations in Asia and in the rest of the world were measured in Riyadh, Saudi Arabia, with concentrations of 640 µg m^{-3} and 370 µg m^{-3} for PM10 and PM2.5, respectively. The range of PM10 concentrations was very broad, ranging from about 30 µg m^{-3} at locations in Australia and Japan to 150 µg m^{-3} in Taipei, Taiwan.

3.4 North America

Monitoring of fine particles is well established in North America, particularly in the United States. Since the PM10 air quality standards were established, intensive PM10 monitoring, particularly in urban areas, was set up. PM2.5 monitoring stations still need to be implemented throughout the country (Tyson, 1997; Kaiser, 1998). Many studies on monitoring of fine particles have been undertaken in North America, and only a general picture from the US is given. An overview on receptor model applications in the US is shown in Table 7.

Brook *et al.* (1997) measured PM10, PM2.5, acidity, and sulfate at urban and rural locations in Canada in the context of health effect studies. The mean PM10 and PM2.5 concentrations at the sites were 11 – 31 µg m^{-3} and 7 – 19 µg m^{-3}, respectively. The inter-site correlations were highest for SO_4^{2-}, followed by PM2.5 and H^+, and were lowest for PM10-2.5. The corresponding H^+ and SO_4^{2-} concentrations were between 4 and 30 nmoles m^{-3} and 21 – 71 nmoles m^{-3}. Ammonium was the dominating cation, sulfate the dominating anion. PM2.5 and PM10 showed a high temporal correlation, but the coarse fraction (PM10-2.5) showed a low temporal correlation to other particulate measures. PM10 and PM2.5 concentrations were highest at the urban sites and in areas with higher SO_4^{2-} concentrations. H^+ concentrations were highest at the Atlantic coast, SO_4^{2-} concentrations were highest in southern Ontario.

US-EPA (1996b) extensively reviewed the state of the art on the monitoring of PM10, PM2.5 and their chemical composition in the United States in the context of the preparation of the update of air quality guidelines in the US. Urban sites have been intensively monitored with the AIRS network since the mid/late 1980s. In the IMPROVE/NESCAUM networks, non-urban sites have been monitored in a much smaller number. Between 1988 and 1994, a substantial decrease in PM10 concentrations was found, in the urban sites usually 20 – 24 %. This decrease was more pronounced in the western regions (23 - 37 %) than for the Northwest (18 - 19 %) and for the industrial Midwest (12 – 19 %). For both, the fine and coarse fraction of PM10, a decrease was generally found. The ratio of PM2.5 to PM10 at urban sites usually averaged 0.55 – 0.6. The PM2.5 to PM10-2.5 ratio showed a much higher spatial and temporal variability. In summer, the seasonal profile of PM10 had a peak for urban and non-urban sites and this was usually associated with PM2.5 peaks. Daily average PM10 concentrations at urban sites usually were between 10 - 60 µg m^{-3}. Natural background levels, excluding all anthropogenic sources in the US and elsewhere, were 4 – 11 µg m^{-3} and 1 – 5 µg m^{-3} for PM10 and PM2.5, respectively.

Table 7 Selected receptor modeling studies from the United States

Location	**Granite City, Illinois**	**Southern California**	**Philadelphia**	**California**	**Bullhead City, Arizona**	**San Joaquin Valley, California**	**Rubidoux, California**	**St. Louis, Missouri**	**Shenandoah Valley, VA**	**Boston, MA**
Source	Glover *et al.*, 1991	Watson *et al.*, 1994	Dzubay *et al.*, 1988	Schauer *et al.*, 1996	Gertler *et al.*, 1995	Chow *et al.*, 1992a	Chow *et al.*, 1992b	Alpert and Hopke 1981	Stevens *et al.*, 1984	Thurston and Spengler 1985
Year of sampling	1986-87	1987		1982	1988-89	1988-89	1988	1975-77		
Size fraction	PM10, PM10-2.5, PM2.5	PM10 (PM2.5)	PM10	Organic fine aerosol, PM2	PM10	PM10	PM10	PM2.4, PM20-2.4	PM2.5	PM2.5, PM15-2.5
Method(s)	Factor analysis, chemical mass balance, wind trajectory		Chemical mass balance, multiple linear regression, wind trajectory receptor models	Receptor model based on organic compounds	Chemical mass balance receptor model	Chemical mass balance receptor model	Chemical mass balance receptor model	Target transformation factor analysis	Factor analysis	Principal component analysis
Major sources	PM2.5: regional sources; PM10-2.5: local industries	In summer: suspended dust; in fall: primary motor vehicle exhaust, secondary ammonium nitrate	Sulfate (49-55 %), crustal matter (17-24 %), vehicle exhaust (4-6 %), stationary sources (<5 %), sulfate mainly from coal and oil fired power plants	PM2: primary (43-64 %): diesel engine exhaust, paved road dust, gasoline-powered vehicle exhaust, food cooking and wood smoke; secondary (36-57 %): sulfate > nitrate > ammonium	Geological dust (80 %), primary motor vehicle exhaust (17 %), secondary ammonium sulfate (4 %)	Primary geological material (54 %), secondary ammonium nitrate (15 %), primary motor vehicle exhaust (10 %), primary construction (8 %)	Road and wind blown dust (52-53 %), secondary ammonium nitrate (23-26 %), primary motor vehicle exhaust (7-11 %), secondary ammonium sulfate (6-8 %), lime and gypsum (5-8 %)	PM2.4: sulfate (84 %), steel (7 %), motor vehicle (6 %), smelters (2 %); PM20-2.4: lime stone (39 %), soil (31 %), sulfate (11 %), soil/fly ash (10 %), steel (5 %)	Coal fired power plants, sulfate and nitrate	Coal combustion 40 % of PM2.5 and 25 % of PM15-2.5

Sulfate, particularly $(NH_4)HSO_4$ or $(NH_4)_2SO_4$, was the largest contributor in the eastern United States, while in the western United States nitrates and organics were relatively more abundant. Sulfate contributed more to PM2.5 than to PM10, soil particles or minerals dominated PM10-2.5. Particle strong acidity was spatially relatively evenly distributed, with some neutralization by ammonia in large urban areas. Fine particles tended to be acid, coarse particles basic. Number concentrations were dominated by ultrafine particles; they ranged from 1000 cm^{-3} up to 100,000 cm^{-3}. Metals were concentrated in the ultrafine particles, particularly those volatilized during combustion.

3.5 Summary of the Available Observations

PM10 mean concentrations were typically 10 – 80 µg m^{-3} around the world. 10 - 20 µg m^{-3} are found at remote sites with no local sources. More than 50 µg m^{-3} were found at heavily polluted sites with high traffic density and/or local sources in the surroundings. In heavily polluted cities of southern Europe, Latin America and Asia, e.g. Mexico City, average PM10 concentrations can reach 100 µg m^{-3} and more. Single measurements may even reach much higher levels.

PM2.5 mean concentrations were between 7 and 80 µg m^{-3}. High levels were found in the winter during coal heating periods and during dry seasons, in large cities of the South, and at locations close to streets with a high traffic density. Daily concentrations can reach several hundred µg m^{-3} and single 1-hour measurements even higher.

A seasonal variation of PM10 ambient levels was not always obvious. In some countries, enhanced particle emissions during the winter heating period were seen, e.g., in Italy, Eastern Europe and Switzerland. In Brisbane, Australia, higher PM2.5 levels were associated with meteorological conditions of more westerly winds transporting inland crustal matter. In the UK, the coarse fraction increased relative to the fine fraction during the summer due to wind blown dust. In other countries, no distinct seasonal variation was obvious, e.g., in Sweden. In general, the seasonal variation is not as pronounced as for other air pollutants.

PM10 and PM2.5 concentrations were elevated close to main roads compared to background sites. The elevation was attributable to elemental carbon from diesel exhaust and suspended road dust.

PM10 trends were decreasing in the last decade, both for emission and ambient levels, in the developed and in the developing world. After abatement of SO_2 emissions started some years ago, particulate sulfate concentrations started to decline. For particulate nitrate, the trend remained increasing even in recent years.

The coarse fraction (PM10-2.5) is usually of similar magnitude as the PM2.5 concentration. The prevalence of local sources, with either a dominating fine or coarse fraction, may shift this ratio. The most common ratio of PM2.5 to PM10 is 0.60. Seasonal variation may also shift this ratio substantially, i.e., the enhanced formation of secondary particles in summer will result in a higher PM2.5/PM10 ratio. Table 8 gives an overview on relative ratios of airborne particles.

Table 8 Relative ratios of PM2.5, PM10 and TSP

Location	**Birming-ham UK**	**Italy**	**Switzerland**	**Netherlands**	**Mexico City**	**Birming-ham UK**	**Germany**
Source	Harrison *et al.,* 1997	D'Innocen-zio 1998	Monn *et al.,* 1997	Janssen *et al.,* 1997	Andrade *et al.,* 1993	QUARG 1996	Kainka *et al.,* 1997
PM10/ TSP		0.87	0.57-0.74				
PM2.5/ PM10	summer: 0.50, winter: 0.80	0.58		0.57		0.48-0.70	0.70-0.80
PM10-2.5 / PM2.5		0.81					0.25-0.50
PM2.5/ TSP					0.40-0.60		
Details		urban air				Monthly averages Jan. – June 95	

Within the size range of 0.01 - 2.5 µm, Tuch *et al.,* 1997 reported about the dominating role of the size range 0.1 – 0.5 µm for the mass concentration. Urban aerosols usually show a bimodal mass distribution with a maximum around 0.2 – 0.3 µm for the coagulation mode and a maximum around 8 µm for the sedimentation mode. A minimum is usually measured around 1 – 2 µm, see Figure 1 (Finlayson-Pitts and Pitts, 1986, p.736). We have plotted the particle mass distribution of Bombay (Sharma and Patil, 1992), Mexico City (Salazar *et al.,* 1992), Ponte, Puerto Rico (Infante *et al.,* 1990) and Jakarta (Zou and Hooper, 1987) and found a minimum around 2 µm. Figure 4 shows the size specific particle mass concentration. Here, the coagulation mode and the sedimentation mode did not have distinct maxima. This shows that the distinction between coagulation mode and sedimentation mode particles reported in the literature can also be seen here.

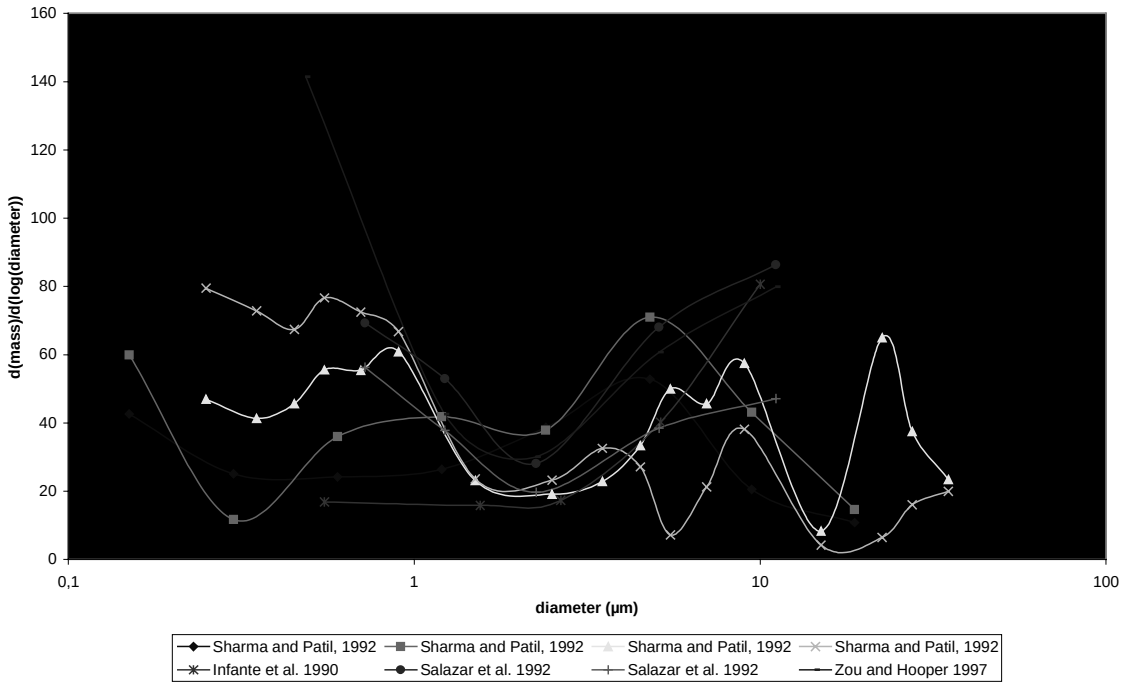

Figure 4 Size distribution of samples from Asia and Latin America

Secondary sulfate particles were more abundant in the fine fraction than in the coarse fraction, whereas secondary nitrate does not show enrichment in the fine fraction. The secondary sulfate particles showed a seasonal profile with higher levels during summer related to enhanced secondary formation processes because of higher temperature and humidity. In contrast, the sulfate concentrations measured in Japan showed a minimum in autumn. Secondary nitrate particles do not show a pronounced seasonal variation.

Elemental carbon was highest in winter and lowest in summer. A similar pattern was found for organic carbon, but not as pronounced as for elemental carbon.

Table 9 summarizes the major components of fine particles. Typically, PM10 contains more than 10 percent of sulfate, organic carbon, and elemental carbon and less than 10 percent of nitrate, chloride, ammonium, and H^+. In the coarse fraction, soil dust and sea salt are relatively more abundant than in the fine fraction. The opposite is true for the combustion products and secondary particles.

Table 9 Major components of fine particles, in %

Component	organic carbon OC	elemental carbon EC	SO_4^{2-}	NO_3^-	Cl^-	NH^4_+	H^+
Mass percent	12-20	10-15	17	4-6	2	3-6	<1

4. Modeling and Source Identification

For fine particle modeling, two approaches have been used: receptor models and dispersion models. For receptor models, detailed information on the composition of ambient particles as well as on the composition of relevant sources is needed. Usually, no information on the meteorology and the spatial distribution of the sources is used. With multivariate statistical analysis, the fingerprints of ambient particles and of the sources are compared and the contribution of the sources investigated. Receptor models have been extensively applied for PM10 studies in the United States. Table 7 gives an overview on the scope and the results of the studies from the US, Table 10 on those from other countries. Because receptor models assume that fractionation and transformation processes are not relevant during transport, the pollutant pattern does not change during transport and dispersion, secondary particle formation is difficult to introduce or to be taken into account. This gives restrictions against the use of receptor models for ultrafine particles and even to some extend for PM2.5. The introduction of some information on wind trajectories and the spatial distribution of the sources may help to address this issue. Figure 5 gives an overview on the source-exposure relationship for the modeling of fine particles.

For dispersion modeling, detailed information on the spatial and temporal distribution and on the meteorology is needed. Data on the ambient levels is also needed for the validation of the model. Episodic models and long-term models are two distinct classes of dispersion models (Seigneur *et al.,* 1997). Episodic models have a detailed treatment of chemical reactions introduced but cover a time period of only few days, whereas long-term models introduce a simplified chemistry but cover longer time periods. The secondary formation of fine particles is covered with varying extent in the dispersion models. The secondary sulfate formation is generally included. The secondary nitrate and the secondary organic aerosol formation have been introduced in several dispersion models. Both regional and local models exist. As model output, usually one particle size range PM10 or PM2.5 is given. Because detailed information on gaseous pollutants, particularly SO_2, NO_x, VOC and ammonia, is needed for appropriate modeling of the secondary formation processes, it is advantageous to introduce a fine particle module into an existing model of gaseous pollutants. For a detailed discussion on fine particle dispersion and receptor modeling, see Seigneur *et al.,* 1997.

Emission inventories are a first and important step in obtaining an overview on the sources of fine particles and their contribution to ambient levels. The most intense activities have been undertaken in the United States. A national inventory for fine particles has been published (US-EPA, 1997). In Europe, TNO published a country-by-country inventory for PM10, PM2.5, and PM0.1 (Berdowski *et al.,* 1997). Only very few national and local inventories are available in Europe. In the UK, a national as well as several local inventories for PM10 were compiled (Salway *et al.,* 1996; Buckingham *et al.,* 1997a; Buckingham *et al.,* 1997b; Buckingham *et al.,* 1997c). In Germany, a national PM10 inventory is being compiled at present from the state authorities for stationary sources (Hoffmann, 1998). In Sweden, Wallin (1998) has prepared a PM10 inventory for the county of Stockholm. Fine particle emission inventories were not available for Latin America, Asia and Africa. Table 11 outlines the emission inventories

for airborne fine particles available. For a detailed discussion on emission inventories, emission factors and their results, see Paounova (1998).

Table 10 Receptor and dispersion modeling studies from various countries

	Santiago de Chile	**Sao Paulo**	**Mexico City**	**Central Taiwan**	**Norrköping, Gothenburg**
Source	Rojas *et al.*, 1990	Andrade *et al.*, 1993	Vega *et al.*, 1997	Chen *et al.*, 1997	Bringfelt *et al.*, 1997
Year of sampling	1987	1993	1989-90	1993-94	
Size fraction	PM2.5, PM15-2.5	PM2.5, PM15-2.5	PM2.5	PM2.5, PM10-2.5	PM10
Method	Absolute Principal Factor Analysis	Absolute Principal Components Analysis	EPA CMB7 receptor model	EPA CMB7 receptor model	MATCH model
Major sources	PM2.5: secondary sulfate (49%), wood-burning/car exhaust (26%), residual oil combustion (13%); PM15-2.5: soil dust (74%), soil dust and industrial release (13%), secondary sulfates (9%)	PM2.5: residual oil and diesel (41%), soil dust (28%), industrial emissions (13%); PM15-2.5: soil dust (59%), industrial emissions (19%), sea-salt (14%), oil burning (8%)	Pre-catalytic converter gasoline vehicles (36-41%), secondary aerosols (20-23%), oil refineries (13-18%), re-suspended dust (10-15%)	PM2.5: agricultural wastes (31-39%), sulfates (16-26%), coal and oil combustion (11-19%), diesel exhaust (7.5-12%); PM10-2.5: soil dust and (paved) road dust (21-42%), burning of agricultural wastes (31-40%), coal and fuel oil combustion (7-27%), marine aerosols (3-11%)	

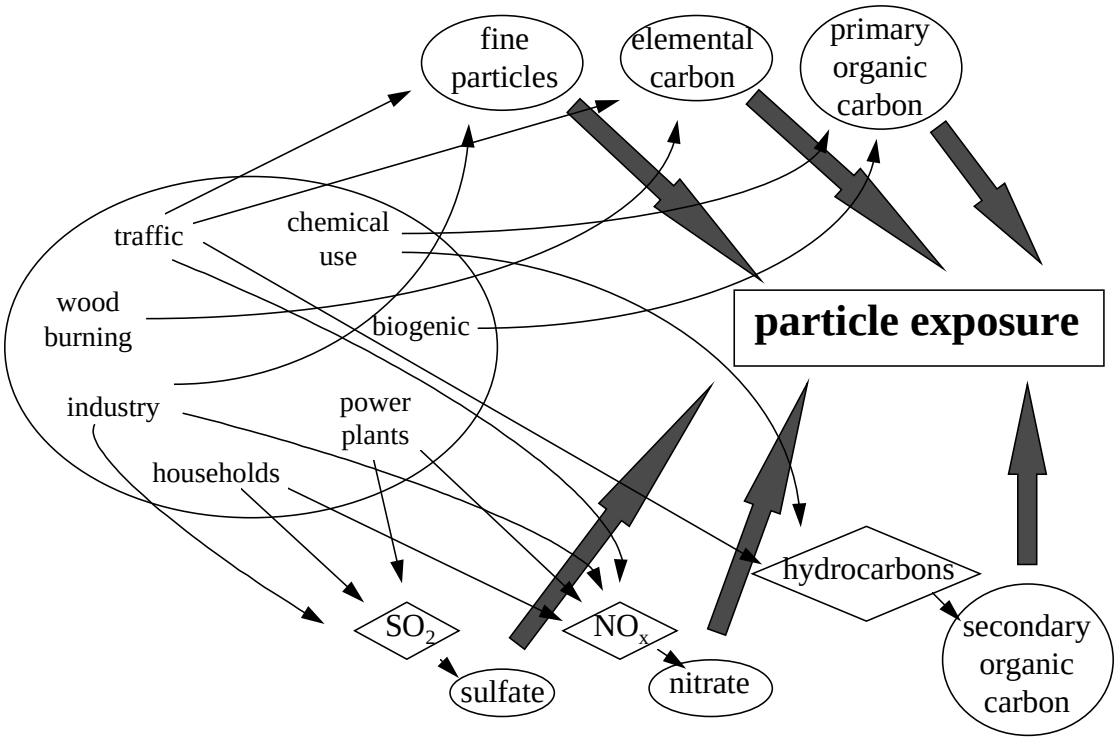

Figure 5 Structure of source-receptor modeling of fine particles

To some extent also single source types have been monitored. Emission factors may be obtained thereof. It remains unclear to what extent the results of these measurements can be extrapolated to other countries and continents and whether these early measurements still reflect the current situation.

The source apportionment from emission inventories and receptor models need to be considered with some caution. Few measurements of fine particles even for major sources have been made in recent years. Often, measurements from the early 1980's are used or measurements of total suspended particles are extrapolated to the relevant size fractions. But the same holds true for results of receptor models, where source compositions are usually taken from the literature without having measured the local sources. It would therefore not be surprising if the focus of relevant sources need to be adjusted in the coming years.

The source contributions to PM10 are usually heavily dependent upon local and regional conditions. The major sources on national level may be different from those on a local level. Sources which make a substantial contribution to annual emissions may be different from those relevant during high pollution episodes.

Table 11 Overview on emission inventories

Region	**Europe, country by country level**	**United Kingdom**	**London**	**Stockholm**	**USA**
	TNO 1997	AEA 1996	LRC 1997	Wallin 1998	US-EPA 1997
Year	1990, 1995	1970 – 1994	1995	1995-96	1900-96
Compound, size range	PM10, PM2.5, PM0.1	PM10 and other air pollutants	PM10 and other air pollutants	PM10, no secondary particle formation	PM10, PM2.5
Categories		UN-ECE categories	General coverage		General coverage
Major sources	PM10: power generation, production processes, road transport, small combustion sources; PM2.5: power generation, production processes, road transport; PM0.1: road transport, production processes, power generation	For 1994: road transport (26 %), non-combustion sources (25 %), industrial combustion (18 %), public power (15 %), commercial residential and institutional combustion (13 %)	Heavy goods vehicles (50 %), cars and taxis (15 %), light goods vehicles (6 %), buses (6 %), waste treatment and disposal (5 %), industrial processes (4 %), public power (4%)	Wood combustion and residential heating (55 %), other types of heating and industry (19 %)	In 1996: fugitive dusts (89 %): unpaved roads, natural sources, agricultural crops, construction; non-fugitive dusts (11 %)
Comments, details	Without natural sources				

5. Conclusions

Ambient levels of fine particles in the environment cause adverse human health effects. Fine particles have been associated with increased mortality and pulmonary and cardiovascular diseases. The majority of epidemiological studies have shown this association for various countries on different continents, for regions with different climatic conditions, for varying levels of gaseous co-pollutants, and for high as well as for low levels of fine particles. No threshold was commonly found and a linear dose-response relationship seems appropriate. Even though this association is still under debate in the scientific community, the epidemiological evidence is reasonably strong. No particle size range has yet been clearly identified as being primarily associated with the adverse health effects and no general biological mechanism has been established. There are indications that the fine fraction (< 2.5 µm) is more likely related to adverse health effects than the coarse fraction (2.5 µm – 10 µm). Seaton *et al.* (1995) hypothesized that ultrafine particles (< 0.1 µm) are the fraction most responsible for health effects.

In this survey we have shown that concentrations of fine particle are high close to main roads and that road transport and particularly diesel exhaust are a major source of fine particles in urban areas, that children and adults living close to motorways experience more respiratory diseases, and that a biological mechanism for the adverse effects of diesel exhaust has been identified. We therefore hypothesize that road transport and particularly diesel exhaust is one, if not the main, cause for excess mortality and morbidity associated with fine particles in urban areas.

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