

Particles in the ambient atmosphere

by Hans Areskoug

Airborne particulate matter (PM) represents a broad class of chemically and physically diverse substances. The principal defining feature of airborne PM is that it consists of discrete particles in the condensed (liquid or solid) phase spanning several orders of magnitude in size, from molecular clusters of a few nanometers in diameter to coarse particles of about 100 micrometers.

Airborne particles can be either primary or secondary in nature. Primary particles are emitted directly into the atmosphere either by natural or anthropogenic processes. Secondary particles have a predominantly man-made origin and are formed in the atmosphere from the oxidation of sulfur dioxide (SO₂), nitrogen dioxide (NO₂), and volatile organic hydrocarbons and subsequent reactions.

In most European countries, industrialization and a high volume of traffic mean that anthropogenic sources dominate, especially in urban areas. The most significant sources are road traffic, power plants, combustion sources (industrial and residential), industrial fugitive dust, loading and unloading of bulk goods, mining activities, and, in some local instances, construction and demolition activities. The main natural sources are sea spray and suspended soil dust.

The major chemical constituents of airborne particles are sulfates, nitrate, carbonaceous compounds (both elemental and organic carbon compounds), acids, ammonium ions, metal compounds, water, and crustal materials.

Physical and chemical characteristics

Particle-size distribution

Size is one of the most important parameters in determining the properties and fate of atmospheric particles. The atmospheric deposition rates of particles, and, therefore, their residence time in the atmosphere is strongly dependent on particle size. Size also influences the particle deposition pattern in the lungs. Light scattering is a strong function of particle size. The scattering efficiency is greatest for particles in the size range 0.3–1.0 μm , and it decreases rapidly for particles outside this range. Therefore, the number of particles and their size distribution have a large effect on atmospheric visibility. Particles suspended in the atmosphere also affect the earth's energy budget, and thus the climate, by increasing the reflection of solar radiation back to space and indirectly by affecting the formation of clouds (1).

Although atmospheric particles are often not spherical, their size is described by an "equivalent" diame-

ter. The equivalent diameter of a particle is the diameter of a sphere having the same physical behavior as the particle in question. The aerodynamic diameter is often used to describe the size of particles. It is defined as the diameter of a spherical particle with the same settling velocity as the particle in question but with a density of 1 g/cm³.

An important feature of atmospheric aerosol size distributions is their multimodal nature. Mass (and volume) distributions are almost always bimodal, with a minimum between 1 and 3 μm (figure 1). The particles that are mostly larger than this minimum are termed coarse. Those that are mostly smaller are termed fine.

Particles are designated as primary if they are emitted directly into the air as particles or as vapors that condense to form particles without a chemical reaction. Secondary particles are formed following a chemical reaction in the atmosphere, which converts a gaseous precursor with a low enough vapor pressure.

The crushing, grinding, and abrasion of surfaces that break large pieces of material into smaller pieces form the coarse mode particles. They are then suspended by the wind or by anthropogenic activity. Energy considerations generally limit the break-up of large particles to a minimum size of about 1 μm . Mining and agricultural activities are examples of anthropogenic sources of coarse mode particles. Fungal spores, pollen, and plant and insect fragments are examples of natural bio-aerosols suspended as coarse particles.

The number distribution of the fine-mode particles reveals three modes (figure 1). The fine-mode particles are originally formed by the conversion of gases to

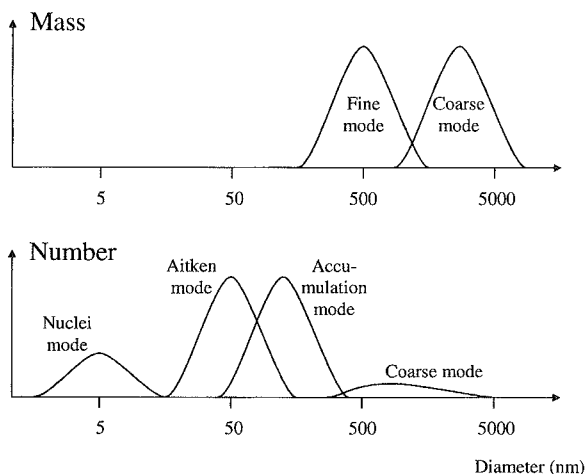


Figure 1. Conceptual description of the particle-size distribution of the ambient aerosol. The relative size of the modes gives the approximate relation between the different modes.

particles. Nuclei-mode particles are the smallest, a few nanometers to 10 nanometers in diameter, and are formed by gas phase chemistry and changes in temperature and relative humidity. Nuclei-mode particles sometimes occur at high concentrations in limited areas. Next in size is the Aitken mode between 20 and 80 nm in diameter. The source of the Aitken particles is not well known. The general assumption is that these particles originate either from primary sources or by growth of nuclei particles by coagulation or by the condensation of gases. Aitken-mode particles grow, through

Table 1. Properties of fine and coarse mode particles. Adapted from Wilson & Suh (2).

	Fine mode ($<2.5 \mu\text{m}$)	Coarse mode ($>1.0 \mu\text{m}$)
Formed from	Gases	Large solids or droplets
Formed by	Chemical reactions	Mechanical disruption (eg, crushing, grinding, abrasion of surfaces)
	Nucleation	Evaporation of sprays
	Condensation	Suspension of dust
	Coagulation	
	Evaporation of fog and cloud droplets in which gases have dissolved and reacted	
Composition	Sulfate, sulfate ion (SO_4^{2-})	Resuspended dust (eg, soil dust, road dust)
	Nitrate, nitrate ion (NO_3^-)	Coal and fly ash
	Ammonium, ammonium ion (NH_4^+)	Metal oxides of crustal elements (silicon, aluminum, titanium, iron)
	Hydrogen ion (H^+)	Calcium carbonate, sodium chloride, sea salt
	Elemental carbon	Pollen, mold, fungal spores
	Organic compounds (eg, polycyclic aromatic hydrocarbons, PNA).	Plant or animal fragments
	Metals (eg, lead, cadmium, nickel, copper, zinc, manganese, iron)	
	Particle bound water	
Sources	Combustion of coal, oil, gasoline, diesel and wood	Resuspension of industrial dust and soil tracked onto roads and streets
	Atmospheric transformation products of nitrogen oxides, sulfur dioxide and organic compounds including biogenic organic species (eg, terpenes)	Suspension from disturbed soil (eg, farming, mining, unpaved roads)
	High temperature processes, smelters, steel mills, etc	Biological sources
		Construction and demolition, coal and oil combustion.
		Sea spray
Atmospheric life-time	Days to weeks	Minutes to hours

liquid-phase chemistry in clouds, to the size range called the accumulation mode, which is 100 to about 1000 nm. These particles are active in forming cloud droplets and are therefore removed from the atmosphere by precipitation.

Fine-mode particles can be of primary or secondary origin. The dominant anthropogenic fine-mode primary particle source is the combustion of coal, gasoline, diesel, and wood. Atmospheric transformation products of SO_2 , NO_2 , and organic compounds form secondary fine-mode particles. The source gases can be anthropogenic or natural.

The gas-to-particle conversion processes in a stack or a tail pipe of a car are similar to those in the atmosphere. The particle-size distribution in the atmosphere is therefore fairly stable despite vastly different source types.

The fine- and coarse-mode particles have substantially different properties, as they are formed in different processes (and are different sizes). A comparison of the two modes is given in table 1.

Generally, particles in the Aitken and accumulation mode dominate the number concentration of particles, while the particles in the accumulation and coarse mode dominate the mass. As the coarse mode is usually dependent on wind speed, ground conditions, and other dust-generating activities, its mass concentration can vary strongly. Another parameter of possible interest is the particle surface. The particles with the largest total particle surface area are those in the accumulation mode.

It should be noted that there is no general consensus among aerosol physicists on the subdivisions of fine-mode particles. Sometimes the nuclei mode, as defined in this report, and the Aitken mode are grouped together and named the nuclei mode. The mode denoted nuclei in this report are then called ultrafine nuclei.

In the terminology of health scientists ultrafine is often used to characterize any size distribution with a mass mean diameter below $0.1 \mu\text{m}$. Ultrafine particles, in reference to health effects, are closely related to the nuclei and Aitken mode. In this document ultrafine is used in its biological context.

Chemical composition

The processes that form the particles in the different modes also affect the chemistry and morphology of these particles.

The smallest particles (ie, nuclei mode) are believed to be formed by the bimolecular nucleation of gaseous sulfuric acid and water vapor. It is likely that gaseous ammonia is involved in the nucleation process, and it neutralizes the particles at least to some extent. However, it is probable that other gases (eg, organic) can also form nuclei particles. The composition of Aitken-mode particles is not well known due to analytical difficulties

and the poor theoretical understanding of the source of this mode. However, indirect methods indicate that the composition is similar to that of accumulation-mode particles (3). The chemical composition of accumulation-mode particles has been investigated in numerous studies. The composition of the urban and nonurban continental fine-particle mode is roughly the same (table 2). Sulfate and carbon compounds make up about 65% of the mass, and ammonium and nitrate about 15%. The carbon compounds are more abundant in urban aerosol (40%) than in the nonurban aerosol (30%), while the opposite is true for sulfate. Only 10% to 20% of organic carbon compounds have been identified in terms of molecular structure (5). Identified compounds include n-alkanes, n-alkanolic acids, aliphatic dicarbocyclic acids, diterpenoid acids, polycyclic aromatic hydrocarbons, and the like (6).

The main feature of the fine-mode particle composition in remote marine regions is the large fraction of crustal and marine components such as silicon, chlorine, calcium, iron, and undetermined compounds. The fraction of elemental carbon, one of the best tracers of anthropogenic emissions, is very low.

In contrast, major components of coarse particles are aluminosilicates and other oxides of crustal elements (eg, iron, calcium) in soil dust; fugitive dust from roads, industry, agriculture, construction and demolition; fly ash from the combustion of oil and coal; and additional contributions from plant and animal material.

Particle size as a function of relative humidity

The interaction between particles and water vapor is important in the atmosphere and has thus been investigated in several studies (3, 7). This interaction is also important in the human lung.

The amount of water absorbed by particles is related to relative humidity in the atmosphere, as well as in the respiratory system. The theoretical growth ratio of sodium chloride particles larger than 100 nm is 5 to 6 when the relative humidity is above 99.5% (figure 2). The smaller the dry size of particles, the lower the growth ratio. The theoretical growth ratios have been confirmed in experiments simulating the humidification process in the human lung (8).

The lower growth for particles less than 100 nm may be important with respect to the biological effect of water-soluble particles. It implies that hygroscopic accumulation-mode particles are more diluted when exposed to humidification in the lungs than nuclei- and Aitken-mode particles are.

The growth of a particle in humid air can also be used as an indirect measurement of the chemical composition of a particle (ie, how much hygroscopic material it contains). In the ambient accumulation mode, two

types of particles are observed (9, 10). One type adsorbs very little water when humidity increases, while the other can adsorb considerable amounts. The less hygroscopic particles grow only by about 5% in diameter at 85% relative humidity, and therefore these particles evidently contain about 95% hydrophobic material, probably soot and organic components. The more hygroscopic particles grow about 40% at 85% relative humidity, and this phenomenon indicates that the particles are predominately inorganic salts such as sulfates, nitrates, and chlorides. However, the growth is far from what is expected for pure salts (see the preceding text), probably because a significant fraction of hygroscopically inactive material is mixed with the inorganic salts. The less hygroscopic particles are more abundant, up to 70%, in urban areas, while in remote regions the relation between the two types is often reversed.

The different growth of the two particle types in a humid environment, like the human lung, leads to different aerodynamic properties, and thus the two particle types deposit in different parts of the respiratory system.

Table 2. Average chemical composition (%) of fine mode particles (<1 μm) in different regions of the world. Adapted from Heintzenberg (4). [C(e) = C (elemental), C(o) = C (organic), NH_4^+ = ammonium ion, NO_3^- = nitrate ion, SO_4^{2-} = sulfate ion]

Region	C(e)	C(o)	NH_4^+	NO_3^-	SO_4^{2-}	Other
Remote ^a	0.3	11	7	3	22	56
Nonurban continental	5	24	11	4	37	18
Urban	9	31	8	6	28	18

^a Mainly data from islands and ship cruises in the Atlantic and Pacific oceans.

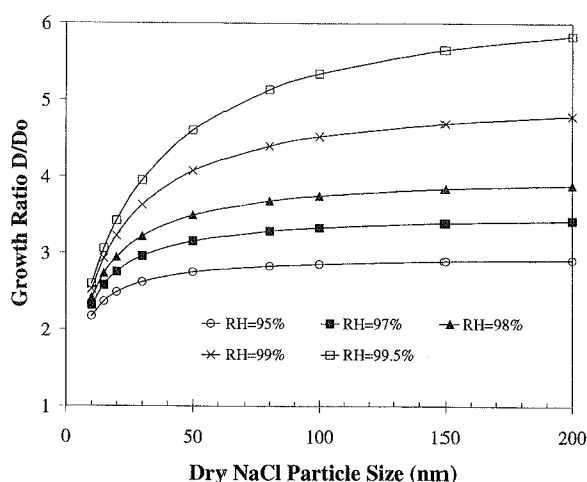


Figure 2. Theoretical predictions of growth of sodium chloride (NaCl) particles at relative humidity between 95% and 100%.

Measurement methods

PM, unlike other air pollutants, tends to be classified by the measurement method used. Thus PM₁₀, PM_{2.5}, black smoke (BS), and total suspended particles (TSP) are all terms used to characterize the concentration of airborne PM, and they give an indication of the measurement method used. These terms and the health-related sampling conventions formulated in an international standard by the International Organization for Standardization (ISO) are defined below (11) as follows:

Inhalable fraction (IF): the mass fraction of total airborne particles that is inhaled through the nose or mouth. For ambient atmospheres the fraction is given by:

$$IF = 0.5 \cdot [1 + \exp(-0.06 \cdot D)] + U \cdot 2.75 \cdot \exp(0.05 \cdot D) \cdot 10^{-5} \quad (U < 10),$$

where D is the aerodynamic diameter (m) of the particle and U is the wind speed (m/s).

Thoracic fraction: the mass fraction of inhaled particles that penetrate the respiratory system beyond the larynx. As a function of total airborne particles, it is given by a cumulative lognormal curve, with a median of 10 µm and a geometric standard deviation of 1.5.

Respirable fraction: the mass fraction of inhaled particles that penetrate to the alveolar region of the lung. As a function of total airborne particles, it is given by a cumulative lognormal curve, with a median of 4 µm and a geometric standard deviation of 1.5.

"High-risk" respirable fraction: the respirable fraction for the sick and infirm or children. As a function of total airborne particles, it is given by a cumulative lognormal curve, with a median of 2.5 µm and a geometric standard deviation of 1.5.

Total suspended particulate (TSP): a term describing the gravimetrically determined mass loading of airborne particles, most commonly associated with the use of the United States (US) high-volume sampler in which particles are collected for weighing. The sampler has a cut point¹ that varies between 25 and 40 µm depending on wind speed and direction.

PM₁₀: particles measured by a sampler that contains a size fractionator of defined characteristics with a 50% sampling efficiency at an aerodynamic diameter of 10 µm. This measurement includes the fine-mode particles and part of the coarse-mode particles and is an indicator for thoracic particles.

PM_{2.5}: particles measured by a sampler that contains a size fractionator of defined characteristics with a 50%

sampling efficiency at an aerodynamic diameter of 2.5 µm. The cut point is primarily chosen to sample the fine-mode particles, but mimics also the "high-risk" respirable fraction as defined by ISO.

Black smoke (BS): nonreflective (dark) PM, associated with the smoke-stain measurement method. Sampled particles are collected on a filter. The quantity of dust is calculated by converting the light reflectance from the surface of the filter using a calibration curve.

The majority of reliable instruments currently available are for the PM₁₀ fraction. They generally make use of a validated sampling head to select the PM₁₀ fraction, the collected particles being collected for analysis with primarily the following two types of instruments: (i) gravimetric, cumulative samplers in which the amount of particles deposited over a sampling period, normally 24 hours, is determined by weighing and (ii) direct-reading monitors in which the particles are either deposited on a filter, with continuous assessment of the change in a property of the filter due to their presence, or passed through an optical sensing unit. The PM_{2.5} fraction can be measured using the same analyzers if an appropriate sampling head is used.

Gravimetric PM₁₀ samplers

Several gravimetric, cumulative samplers are available. The main difference between them is the flow rate, which ranges from 16.7 to 1130 l/min. The samplers with a high flow rate have the advantage of providing a sufficient sample for both gravimetric and chemical analysis, but they are larger than those with a low flow rate. Gravimetric-type samplers constitute the reference in the United States and are also the proposed European reference.

In the proposed European reference method, the filter is equilibrated for 48 hours before and after the sampling at 20±1°C and a relative humidity of 50±5%. This process is supposed to remove most of the particle-bound water, but may also lead to some loss of volatile compounds, including volatile hydrocarbons and ammonium nitrate. More importantly, the ambient conditions of temperature and humidity that the sample is exposed to both during and after collection (before removal from the sampler) are completely uncontrolled and can lead to additional variability in the amount of PM.

Direct-reading PM₁₀ samplers

The direct-reading samplers normally use a low-flow PM₁₀ or PM_{2.5} inlet. Several different instruments are

¹ When used in the context of sampling, cut point is a term to describe the separation efficiency curve of samplers. The cut point is typically described by the aerodynamic diameter at which the sampler achieves 50% collection efficiency.

available, and they can be categorized into the following three main categories: optical, beta particle, and resonance oscillation.

The optical instruments use the interaction between airborne particles and visible light in a sensing region to determine the amount of particles. As the response is dependent on the size distribution, shape, and refractive index, the instruments require calibration to give results in terms of mass. The calibration only holds if the particle properties do not change, and hence measurements obtained in the ambient atmosphere can be very uncertain.

In the beta-particle instruments, the attenuation of the intensity of beta particles passing through a dust-laden filter is measured. The change in attenuation reflects the amount of particles collected. As the change is not only dependent on the mass of the particles, but also on the elemental composition, the instrument must be calibrated, and hence the results, as for the optical samplers, are uncertain.

TEOM® sampler. The Tapered Element Oscillating Microbalance (TEOM) is the only direct-reading monitor in which the output is directly dependent on mass. The sample air passes a filter on top of the narrow end of a hollow tapered glass tube. As particles collect on the filter, the mass, and thus the natural frequency of oscillation of the tube, changes. The frequency change is directly proportional to the added mass. The sampling inlet including the sensing unit is heated to 30°C or 50°C to drive off any sampled water.

Several researchers have reported that the modification of the aerosol by the elevated operating temperature appears to have a significant effect (loss) on mass concentrations under certain conditions (ie, a high concentration of ammonium nitrate and volatile organic hydrocarbons). Meyer et al (12) co-located a TEOM with a gravimetric PM₁₀ sampler in Mammoth Lakes, in the United States, during a winter heating season (heavy wood stove usage). The regression between the two samplers had a strong correlation ($r^2 > 0.98$), but the TEOM results were 35% and 45% low for operation at 30°C and 50°C, respectively. The negative bias of the TEOM was attributed primarily to a loss of semivolatile organics from the filter. A WESTAR (13) council report summarizes the relationship between TEOM monitors and cumulative gravimetric samplers in at least 10 states in the western part of the United States. The report concludes that, on the average, the TEOM sampler showed 22% lower concentrations than other co-located PM₁₀ samplers at concentrations above 50 µg/m³. On the other hand, several studies (eg, reference 14) have reported a consistent and linear relationship between the TEOM sampler and gravimetric samplers.

As already noted, a loss of volatile PM can occur also with gravimetric filter methods. The loss can vary greatly, due to the uncontrolled conditions during and after the sampling, but it is, in general, lower than for TEOM samplers. Whether the sometimes lower, but well-defined (in terms of sampling conditions) TEOM results are to be preferred before the perhaps more realistic, but also more variable results obtained by the gravimetric filter methods is an open question. The discussion highlights the problems of particle sampling and raises the question of whether the sampling conditions should be tightly controlled in the reference methods as suggested by, for example, Smith et al (15). However, the possible difference between the TEOM sampler and the gravimetric filter methods should be kept in mind when data obtained with the TEOM sampler are used to determine compliance with air quality standards for PM₁₀. These standards are often based on PM₁₀ data obtained with gravimetric filter methods or by the conversion of BS or TSP data to PM₁₀ using conversion factors obtained in a comparison of TSP and BS with gravimetric filter methods.

Other particle-sampling methods

BS sampler. BS is determined in Sweden by a version of the method introduced by the OECD (Organization for Economic Co-operation and Development) in 1964. Air is drawn through an inverted funnel and a few meters of plastic tubing. The cut point of the method is about 4 to 7 µm at low wind speeds, but lower at high wind speeds. The collected particles are deposited on white filter paper. The mass concentration of BS is calculated by converting the light reflectance from the surface of the filter using a calibration curve. Soot particles composed of elemental carbon, found in incomplete fossil-fuel combustion products, typically contribute most to the darkness of the particles. Thus the amount of elemental carbon, but not organic carbon, correlates the best with the reflectance. Other nonblack, noncarbon particles also have optical properties that can affect reflectance, although their contribution is usually negligible.

Since the relative proportion of atmospheric carbon and noncarbon particles can vary greatly from time to time and site to site, the same reflectance can be associated with markedly different amounts of collected particles or even with markedly different amounts of carbon. The calibration curves used in the OECD method are based on a European standard for soot from the time of the introduction of the method. At that time, coal was the predominant fuel used for heating. As the properties of particles have changed during the years depending on changing combustion and cleaning techniques, the mass concentrations given by the OECD method

cannot be compared over time or between different locations without considerable caution (16).

Several studies have found varying (site- and season-dependent) ratios for TSP and PM_{10} to BS. In the European study Pollution Effects on Asthmatic Children in Europe (PEACE), gravimetric PM_{10} samplers and BS samplers were collocated at 28 sites in 10 countries in the winter of 1993–1994 (17). Four Swedish sites were included, two in the southern and two in the northern part of the country. The correlation between PM_{10} and BS measured at the same site was high at most sites, but the median PM_{10} to BS ratio varied from 0.67 to 3.67. The average ratio was 1.97. The ratios varied less, from 2.45 to 3.67, at the Swedish sites.

In another study, results from three collocated BS and PM_{10} samplers at three English sites also correlated well at each site (18). The correlation was stronger in winter than in summer, indicating the larger contribution of black motor-vehicle-derived PM_{10} in winter and the larger contribution of secondary noncarbon PM_{10} in summer. The PM_{10} and BS concentrations were about the same in winter, but during summer the PM_{10} concentrations were systematically higher. The calculated regression coefficients for BS as a function of PM_{10} were also site-specific and, therefore, reflect the contribution of the different sources to the observed concentrations of PM at each site.

Coefficient of haze. A particle sampler for the coefficient of haze (CoH) has been employed for routine monitoring in many cities in the United States. The sampler collects particles with a 50% cut point of about 5 μm . The sampled particles are collected on a filter-paper tape that is periodically advanced to allow accumulation of another stain. The opacity of the stain is determined by the transmittance of light through the deposited material. The results are expressed in terms of optical density or the CoH per 1000 linear feet of air sampled (rather than in mass units). The CoH sampler is somewhat more responsive to noncarbon particles than the BS sampler is, but, again, any attempt to relate the CoH to micrograms per cubic meter requires site-specific calibration by a collocated gravimetric sampler.

Conversion to PM_{10} from other particulate measures

A variety of measures of airborne PM has been used in epidemiologic studies. If the health effects are to be related to a common measure (eg, PM_{10}) conversion factors from other measures have to be applied. Dockery & Pope (19) have suggested conversion factors of $PM_{10} = PM_{15} = BS = PM_{2.5}/0.6 = CoH \times 55 = TSP \times 0.55 = (\text{particulate sulfate}) \times 4$. These factors are used in the section on epidemiologic studies in this document. As already described, conversion factors are both site- and

time-dependent, making them highly variable, especially the factors that convert the optical measures BS and CoH. For example, the assumption that BS concentrations are equal to PM_{10} is based on studies in the United States. This relation does not seem to be valid in Sweden, where PM_{10} are two to four times the BS concentration; see the preceding discussion. The conversion factors for the gravimetric measures ($PM_{2.5}$, PM_{15} , TSP, and particulate sulfate) are probably more accurate in most cases.

Main sources of particulate matter

In comparison with sources of other air pollutants there are two features of PM that increase the difficulty and uncertainty to estimate its emission. The first is that the sources that emit particles not only include fuel use, but also fugitive sources, whose scale and activity rates are poorly known. The other is the varying definitions of PM used in emission estimates. Sometimes the estimates refer to the total amount of suspended particles emitted, sometimes to the PM_{10} particles. Moreover, in most cases, no information is available on the size distribution of the emitted particles. Another difficulty is that emission inventories for particles in general contain no information on sources of secondary particles, which in many areas can make up a significant proportion of ambient concentrations.

Natural sources

On a global scale, several natural sources, such as suspension of soil dust, sea salt, volcanic activities, forest and other wild fires, and an ingress of extraterrestrial particles, are significant sources of particles emitted into the atmosphere.

The action of the wind on dry loose surfaces leads to particles blowing into the air. This phenomenon is most obvious during sand and dust storms in the world's deserts. These processes do, however, extend to other regions of the globe, although their magnitude is reduced where soils are moist and have vegetation cover.

Breaking waves on the sea cause the ejection of tiny droplets into the atmosphere. When the droplets evaporate, they leave mostly coarse, sea salt particles in the air. However, some of the particles are small enough to have an appreciable atmospheric lifetime. All parts of Sweden are influenced to some extent by sea salt particles. Coastal areas are obviously affected the most, especially the west coast of Sweden, where the salinity of sea water is substantially higher than in the Baltic Sea.

Secondary particles are formed naturally (i) by the oxidation of sulfur gases from oceans, wetlands, volcanoes and forest fires, (ii) by the oxidation of nitrogen

oxides (NO_x) produced by soils, forest fires and lightning, (iii) by the emission of ammonia from wild animals and undisturbed soil, and (iv) by the oxidation of hydrocarbons emitted from vegetation (terpenes, waxes) and wild fires.

A comparison of the global natural and anthropogenic emissions of particles is given in table 3. Overall the natural emissions are about 10 times the anthropogenic levels. However, the emission of fine particles is dominated by anthropogenic sources. In the densely populated parts of the Northern Hemisphere, including Sweden, natural sources are less important in comparison with the particles arising from human activities. The concentrations of PM₁₀ in Scandinavia are 10–50 times the estimated natural levels (21).

Table 3. Global emission estimates for major aerosol types in the 1980s. Source: Kiehl & Rodhe (20).

Source	Estimated flux (million tons per year)		Comments
	Fine particles ^a	Coarse particles ^a	
Natural			
Primary			
Soil dust	1000—3000 ^b		Mainly coarse
Sea salt	-	1000—10 000	
Volcanic dust	-	4—10 000	
Biological debris	-	26—80	
Secondary			
Sulfates from bio-genic gases	80—150	-	
Sulfates from volcanic sulfur dioxide	5—60	-	
Organic matter from biogenic volatile organic compounds	40—200	-	
Nitrates from nitrogen oxides	15—50 ^b		Fine and coarse
Total natural	130—435 ^c	2000—23 100 ^{c, d}	
Anthropogenic			
Primary			
Industrial dust, etc	4—130 ^b		Mainly coarse
Soot	5—20	-	
Secondary			
Sulfates from sulfur dioxide	170—250	-	
Biomass burning	60—150	-	
Nitrates from nitrogen oxides	25—65 ^b		Mainly coarse
Organics from anthro-pogenic volatile organic compounds	40—200	-	
Total anthropogenic	275—650	30—200 ^e	
Total	400—1 100	2030—23 300	

^a Coarse and fine size categories refer to mean particle diameter above and below 1 µm, respectively.

^b The sum of fine and coarse modes

^c Includes 50% of the nitrates formed from natural emissions of nitrogen oxides.

^d Includes all soil dust.

^e Includes all industrial dust and all nitrates formed from anthropogenic emissions of nitrogen oxides.

Anthropogenic sources of primary particles

Emissions from mobile sources. All mobile vehicle sources emit particles formed by the incomplete combustion of fuel and lubricating oil. Road transport also generates particles due to the wear of brakes and tires. The produced particles are generally smaller than 10 µm (table 4). Road transport can also generate high concentrations of particles in the vicinity of roads by the resuspension of particles deposited on road surfaces.

Particles form in high-temperature combustion processes in diesel and gasoline engines. Initially only soot particles are formed, while the other compounds are in the vapor phase. The hydrocarbons and, in the case of diesel, sulfate (which results from the fuel sulfur) condense as the temperature drops and form particles with a carbon core and a coating of hydrocarbons and sulfate. The particles grow by coagulation, condensation, and adsorption, predominantly after the gas has left the exhaust pipe, as the residence time is too short and the temperature too high for adsorption and coagulation.

The particle emission from diesel vehicles is more than a hundred times as high as the emission from gasoline-driven cars equipped with a three-way catalyst, while the emission from cars without a catalyst is about 10 times the emission from a catalyst car (table 5). The emission factors are calculated from standardized

Table 4. Size fraction of particulate matter emission from road transport. Source: US EPA (22).

	Mass percentage in size fraction	
	<2.5 µm	<10 µm
Gasoline engines		
Leaded — no catalyst	67	80
Unleaded — no catalyst	70	90
Three-way catalyst	90	97
Diesel engines		
	92	100
Tire wear	39	55
Brake lining	39	98

Table 5. Estimated emission factors for particles for gasoline-fueled light-duty vehicles, with and without a 3-way catalyst (TWC) (23, 24) and for diesel fueled-heavy-duty vehicles using an environmental class 1 (MK1) fuel and a nonclassified diesel fuel (MK3) (25).

	Estimated emission factors for particles (mg/km)
Gasoline passenger cars	
Light-duty without TWC, model 1993	20
Light-duty with TWC, model 1993	2.4
Diesel vehicles	
Heavy-duty vehicle MK1	299
Heavy-duty vehicle MK3	336

driving cycles for diesel- and gasoline-fueled vehicles. The gasoline-fueled vehicles are tested according to the federal test procedure of 1973, the diesel vehicles in a transient driving cycle simulating public transportation within a city area.

Emission factors are dependent on several factors (eg, weather conditions, speed, and engine conditions), and it is therefore difficult to make emission estimates based on them. However, when traffic frequency is taken into account, the annual exhaust particle emissions in Sweden from heavy and light duty diesel vehicles are estimated to be 6600 and 560 t/year, respectively (K Andersson, Swedish EPA, personal communication). The particle emission from gasoline-fueled cars with and without a catalyst is estimated to be 280 and 1360 t/year, respectively.

The movement of vehicles on streets results in the resuspension of road dust. The road dust deposit available for resuspension originates from the mechanical wear of, and dirt on, vehicles (including tire and brake lining wear), debris from loads on vehicles, influx of soil material, and the like. Data from the US Environmental Protection Agency (EPA) indicate that the resuspended dust mainly consists of coarse particles. About 20% of the particles are less than 10 μm in diameter, and about 9% have a diameter of less than 2.5 μm (26).

In Scandinavia, the use of studded tires in winter on paved roads without snow or ice cover may wear the road surface to such an extent that the resulting particle deposit on the road becomes the dominating source of resuspended particles (27). Sanding and salting to prevent slippery roads because of snow and ice are also important sources.

The amount of resuspended particles is thus dependent on many parameters (eg, traffic density and the ratio between the number of cars and buses or trucks, humidity, temperature, precipitation amount and duration of dry periods, frequency of sanding and salting, etc).

A model to calculate the PM_{10} concentrations due to local emissions, including resuspension, has been developed at the Swedish Meteorological and Hydrological Institute (28). The model has been applied to a central street in Stockholm (Hornsgatan, 33 000 vehicles/day, 93% cars) for two winter seasons (1 October—31 March, 1994—1995 and 1996—1997) and one summer (1 April—30 September 1995). The results showed that resuspension was the dominating source of PM_{10} during the winter season and generally accounts for between 20% and 50% during the rest of the year (29). The model was also applied for the winter season 1995—1996 to an area of about 15 $\times$ 20 km, including the central parts of Stockholm. The resuspension of particles contributed, on the average, 15—20% of the total PM_{10} concentration, which is about the same contribution as direct emissions from mobile sources.

It is tempting to try to estimate the total Swedish emission of resuspended particles by extrapolating the findings from Stockholm. However, it is not possible, as the parameters that control resuspension can differ markedly between the regions of Sweden (eg, the frequency of sanding and salting, ice and snow cover on roads, etc). Moreover, the Stockholm results are based on model estimates for the winter. The model findings must be further validated by measurements before they can be used for general assessments of emissions. However, the emission of PM_{10} particles due to resuspension may be of the same order of magnitude as the direct emissions from cars and trucks.

There are several other mobile sources that emit particles. The annual emissions from ships on Swedish territorial waters have been estimated to be 2000 t (30). Cargo ships are responsible for about 15%, ferries for about 23%, and pleasure boats for the rest. The large emissions from pleasure boats are due to the frequent use of 2-stroke engines, which only combust 75% of the fuel.

The emissions from airplanes consist of very small particles (<2.5 μm), soot, and engine wear. The magnitude of the emissions has been estimated from fuel-related emission factors. They are not significant emissions on a national scale, but may be of importance locally. The emissions during landing and take-off in the Stockholm area (including the largest civil airport in Sweden, Arlanda) are estimated to be 26 t/year (29).

Emissions from motorized tools and work machines are not regulated. This is a broad category that includes a variety of domestic, commercial, agricultural and forest sources, such as air compressors, snowmobiles, and lawnmowers. As they are often operated during long periods, the emissions of particles may be significant, especially at road and construction sites. The annual emissions in Sweden are estimated to be 6780 t, almost as much as the direct emissions from diesel and gasoline vehicles (K Andersson, Swedish EPA, personal communication).

Emissions from stationary combustion. The emission of particles due to the use of fossil fuel for heating is very important in the cold season. The emission factors for a residential heating unit and a large central heating unit for a shopping center fueled with oil have been estimated to be 140 and 100 mg of particles per kilogram of fuel, respectively (31). This level is lower than the emission factors for diesel vehicles; see the preceding discussion. The annual Swedish emission from residential heating has been estimated to be 580 t by Hansson et al (21).

Heavy oil is mainly burned in large power plants that are used at high consumption peaks, generally during cold winter days. The annual emissions are thus strong-

ly dependent on the meteorology. The total emission in 1994 is estimated to have been 5000 t, based on an oil consumption of 3.28 million cubic meters and using an emission factor of 1.5 grams of particles per kilogram of fuel (21). The emission from coal and charcoal combustion is estimated to be 1180 t/year (21).

In addition to fossil fuels, wood is burned for residential heating and occasionally in forest fires. Forest fires are significant particle sources on a global level, but is normally of less importance in the humid climate of Sweden. However, emissions from fuel wood for heating are a major source locally, especially during the cold part of the year.

Emissions from wood combustion consist mainly of PM, tar, and volatile compounds. As the combustion is often inefficient, large quantities of particles and organic matter are emitted. The composition of the particles is not known in detail. Tar is made up of a great number of condensable organic compounds, among which the polycyclic aromatic hydrocarbons are an important group. The annual Swedish emission of particles due to wood combustion is estimated to be in the broad interval of 8600–65 000 t (21).

Industrial processes. Industrial emissions can be a significant source of particulate emission in certain areas, depending on the location of the industry and the abatement technology used. Although many studies have been conducted to characterize emissions from industrial sources, for example, steel works, information on small emitters such as metallurgical processes and small factories is more limited. Particles emitted from these sources have been found to be in the size range of 0.5 to some 100 µm, depending on the nature of the source.

For precise emission estimates, detailed information is needed from each emitter. Such information is normally not available, and thus the estimates are very imprecise. Kindbom et al (32) have estimated the particulate emission for 1990–1995 to be 9000 t/year. A large fraction of the particles are likely to be larger than 10 µm.

Dust from construction sources. The amount of dust emission from a construction site depends on the size of the site and the amount of construction activity.

The annual emission of the PM₁₀ fraction of construction dust in the United Kingdom has been estimated to be 500–8300 t (18). The estimate is based on an American study around the construction site of an apartment and shopping center. By taking into account the size of the American study area, the amount of construction in the United Kingdom and the more humid climate in the United Kingdom, the emission range was calculated.

By further extrapolation to Sweden, on the assumption of the climate being about the same and the amount

of construction being proportional to the population (United Kingdom 57 million, Sweden 9 million), the Swedish emissions can be estimated to be 80–1300 t/year. Although this estimate is very rough — one could expect that the colder climate in Sweden than in the United Kingdom would reduce the Swedish emissions to some extent — the estimate shows that construction dust is a minor source on the national level, but may be important locally.

Summary of emission estimates for primary particles

The estimates given are summarized in table 6. They are essentially the same as those given by Hansson et al (21). They differ in that: (i) dust emissions from construction sites are included and (ii) new data on emissions from industrial sources (excluding combustion) are used.

Wood combustion is probably the major source, although the emission estimate is very uncertain. A better estimate requires better knowledge of wood consumption, emission factors, and also factors determining the combustion efficiency (eg, type of stove and condition of the wood fuel).

When the magnitudes of different sources are compared, one should be aware that the emissions from traffic, direct exhaust emissions, dust resuspension, and emissions from residential heating, especially wood combustion, generally occur close to the ground and in areas where people spend much of their time. Thus these

Table 6. Estimates of annual particle emissions in Sweden, 1995. (TWC = 3-way catalyst)

Source category	Particles (t/year)	Comments
Mobile sources		
Road transport		
Engine emissions		
Heavy-duty diesel	6600	
Light-duty diesel	560	
Petrol with TWC	280	
Petrol without TWC	1360	
Resuspension	5–10 000	Probably in the same order of magnitude as engine emissions from mobile sources
Other mobile sources		
Working machines	6780	
Ships	2000	On Swedish territorial waters
Air craft	<100	Insignificant on a national level
Stationary combustion		
Heavy oil	5000	
Domestic oil	580	
Coal and charcoal	1180	
Wood	8600–65 000	
Industrial processes	9000	
Construction	80–1,300	
Total	49 000–112 000	

sources are normally more important than the remaining sources from a health point of view. The strength of the sources can also vary considerably over time. For example, the resuspension of road dust can differ substantially depending on the road surface being wet or dry.

Sources of secondary particulate matter

Secondary PM is an important contributor to suspended particulate mass. Sulfate is formed by the oxidation of SO₂, nitrate by the oxidation of NO₂, ammonium from ammonia and aerosol organic carbon species by the oxidation of a large number of precursors. Thus the formation rate of a substantial fraction of the PM is given by a complex function of both emission rates of precursor gases and the rate of chemical processes in the atmosphere.

To be able to use precursor emission estimates to model the formation of secondary particles, detailed knowledge of the conversion processes is needed. While the mechanism for forming inorganic secondary particles is fairly well known, that for organic secondary aerosol is not well understood.

The emission of the precursor gases SO₂, NO_x, ammonia, and nonmethane hydrocarbons in Sweden in 1995 is estimated to be 94, 355, 61, and 458 thousand tonnes, respectively. These values cannot directly be translated into production rates of particles. While, for example, the chemical oxidation of SO₂ leads quantitatively to the formation of sulfate, the formation of aerosol from the oxidation of nonmethane hydrocarbons is much less because only a small fraction reacts to form particles.

Dry deposition and precipitation scavenging of some of the precursor gases may occur before the gases are transformed to aerosols. In addition, a large fraction of the gases is transported out of Sweden before being oxidized. Likewise, oxidation products formed from precursor gases emitted in countries outside Sweden constitute a considerable fraction of the airborne PM in Sweden.

This "import-export" of airborne PM can be illustrated by model calculations made within the Co-operative Program for Monitoring and Evaluation of the Long-range Transmission of Air Pollutants in Europe

Table 7. Import-export budgets of oxidized sulfur (kt/year) and oxidized and reduced nitrogen (kt/year) for Sweden, average 1985–1994. Source: Barrett et al (33).

Constituent	Emissions	Export	Deposition	Import ^a
Oxidized sulfur	85	65	219	199
Oxidized nitrogen	124	109	120	105
Reduced nitrogen	58	27	80	51

^a Import = deposition in Sweden originating from non-Swedish sources.

(table 7). Sweden is a net importer of oxidized sulfur and reduced nitrogen and a net exporter of oxidized nitrogen, but it exports the major part of the SO₂ and NO_x emitted by domestic sources and about half of the emitted reduced nitrogen.

Long-range transport

The long-range transport of particles can be viewed as a separate source when the importance of Swedish control measures to reduce the particle emissions are being assessed.

The long-range transport of particles to Scandinavia is totally dominated by anthropogenic fine-mode particles, which have an atmospheric life-time long enough to be transported thousands of kilometers. Moreover, the fine-mode generating sources in northern Europe are almost solely anthropogenic. A major fraction of these particles is probably secondary particles.

Long-range transport dominates the concentrations of particles and other pollutants in remote areas of Sweden not influenced by any nearby sources. Persson et al (34) estimated that long-range transport constitutes an average of about 50% of the airborne particles in southern Sweden and, on the average, is the dominating source in the north. Tarrasón & Tsyro (35) estimated that the contribution to the concentration of inorganic PM_{2.5} particles due to sources abroad averages 90% for Sweden. The model results should be treated with caution, as the model did not consider primary particles. Moreover, elemental and organic carbon was not considered by Tarrasón & Tsyro, and it was calculated in a simplified way by Persson et al.

Ambient concentrations

Regular monitoring of the health-related particle sizes (ie, PM₁₀ and PM_{2.5}) in Sweden did not commence until the beginning of the 1990s. Data are still sparse, especially PM_{2.5} data, as the measurements are made mainly in a few large towns, mostly in the southern part of the country. Most of the available data are from measurements at roof level in city centers. Data on the concentrations at street level are primarily from short-term campaigns. The rural, background concentration is monitored at one station 100 km south of Stockholm. Unfortunately, no data on the concentrations in a residential area where wood burning is used for heating is available. A summary of all available data on PM₁₀ in Swedish cities is shown in table 8 (roof level) and table 9 (street level).

Roof level (urban background) PM₁₀ concentrations

The annual mean concentration of PM₁₀ at roof level is 12–20 µg/m³. The 98th percentiles of the daily mean

values are in the range of 30–40 $\mu\text{g}/\text{m}^3$, the maximum daily mean concentrations being in the range of 50–100 $\mu\text{g}/\text{m}^3$. The ratios between the 98th percentile and the mean are 2.3–2.7.

The variation of roof-level PM_{10} concentrations between the cities is strikingly small. The main reason is the strong influence of the long-range transport of particles in southern Sweden. Consequently, the difference between the concentrations at roof level in the towns and the concentrations in rural air is also small. The average concentration at roof level in central Stockholm in 1994–1997 was 15 $\mu\text{g}/\text{m}^3$ compared with 10 at a rural background station about 100 km from the town center. The 98th percentile of the daily means in the city was 36; at the background station it was 30 $\mu\text{g}/\text{m}^3$.

Table 8. Summary of PM_{10} concentrations at roof level in Swedish cities. Data from the local environment and health protection administrations. (Summer = 1 April – 30 September, Winter = 1 October – 31 March, 98%-day, max-day, 98%-hour, max-hour: 98th percentile and maximum of daily and hourly averages, respectively)

Station	Period	Aver age	98%- day	Max- day	98%- hour	Max- hour
Göteborg, Femman	Summer 90	14	..	52	..	..
Malmö, Kalendegatan	Summer 90	8	..	60	..	..
Göteborg, Femman	Winter 90–91	17	..	59	..	..
Malmö, Kalendegatan	Winter 90–91	20	..	90	..	..
Göteborg, Femman	Summer 91	14	..	44	..	..
Malmö, Kalendegatan	Summer 91	7	..	43	..	..
Göteborg, Femman	Winter 91–92	15	..	70	51	211
Malmö, Kalendegatan	Winter 91–92	11	..	80	..	..
Göteborg, Femman	Summer 92	15	..	53	..	..
Malmö, Kalendegatan	Summer 92	6	..	42	..	..
Göteborg, Femman	Winter 92–93	15	..	53	48	260
Malmö, Kalendegatan	Winter 92–93	8	..	116	..	..
Norrköping, Lindö	Winter 92–93	12	28	..	34	58
Göteborg, Femman	Summer 93	15	..	64	..	..
Stockholm, Torkel K	Summer 93	12	..	22	..	..
Göteborg, Femman	Winter 93–94	17	..	65	58	275
Stockholm, Torkel K	Winter 93–94	15	..	34	..	..
Göteborg, Femman	Summer 94	14	..	31	..	..
Malmö, Kalendegatan	Summer 94	26	..	78	..	..
Stockholm, Torkel K	Year 1994	14	38	48	..	..
Göteborg, Femman	Winter 94–95	15	..	45	43	162
Malmö, Kalendegatan	Winter 94–95	12	..	45	..	..
Norrköping, Rosen	Winter 94–95	13	..	43	..	..
Oxelösund, vattentornet	Winter 94–95	13	..	56	..	..
Göteborg, Femman	Summer 95	13	..	50	..	..
Norrköping, Rosen	Summer 95	14	..	42	..	..
Stockholm, Torkel	Year 1995	15	34	..	..	..
Göteborg, Femman	Winter 95–96	17	..	53	..	..
Norrköping, Rosen	Winter 95–96	15	41	..	47	167
Oxelösund, vattentornet	Winter 95–96	16	..	60	..	..
Göteborg, Femman	Summer 96	16	..	55	..	..
Malmö, Kalendegatan	Summer 96	16	..	44	..	..
Stockholm, Rosenlund	Year 1996	16	38	49	..	..
Göteborg, Femman	Winter 96–97	13	..	47	..	..
Malmö, Kalendegatan	Winter 96–97	16	..	59	..	..
Malmö, Kalendegatan	Summer 97	16	..	49	45	97
Stockholm, Rosenlund	Year 1997	15	36	..	..	..
Malmö, Kalendegatan	Winter 97–98	14	..	47	39	100

Street level PM_{10} concentrations

The concentrations at street level in the major cities in southern Sweden vary considerably more; this variation can be explained by varying traffic density and type of street (eg, street canyon versus street without surrounding buildings). The mean concentrations are in the range of 20–40 $\mu\text{g}/\text{m}^3$, the 98th percentile of the daily means being 50–100 $\mu\text{g}/\text{m}^3$. The ratios between the 98th percentile and the mean extend from 2.2 to 3.0. The maximum daily concentrations are in the range of 100 to 150 $\mu\text{g}/\text{m}^3$, and the hourly mean concentrations can be up to 300 $\mu\text{g}/\text{m}^3$.

PM_{10} and black smoke in northern Sweden

Very little data on PM_{10} are available from northern Sweden. However, soot levels in several small municipalities in the inland of northern Sweden in general were about the same as in the major cities in southern Sweden (36, 37). Soot levels in six small northern municipalities in January to March 1990 were substantially elevated during cold periods (38), possibly because of the low dispersion during the stable conditions often prevailing during cold weather. In addition, low temperatures directly influence the emissions of soot (and PM_{10}) through the need for increased house heating. Wood burning for residential heating is widespread in this area

Table 9. Summary of PM_{10} concentrations at street levels in Swedish cities. Data from the local environment and health protection administrations. (Summer = 1 April – 30 September, Winter = 1 October – 31 March, 98%-day, max-day, 98%-hour, max-hour: 98th percentile and maximum of daily and hourly averages, respectively)

Station	Period	Aver age	98%- day	Max- day	98%- hour	Max- hour
Göteborg, Riddargatan	May 94	15	..	37	..	..
Göteborg, Artillerigatan	Winter 94–95	18	..	86	..	..
Stockholm, Hornsgatan	Winter 91–92	42	..	133	..	..
Stockholm, Hornsgatan	Summer 92	27	..	51	..	..
Stockholm, Hornsgatan	April - June 94	38	..	127	129	..
Stockholm, Hornsgatan	April - June 95	35	..	..	112	..
Stockholm, Hornsgatan	April - June 96	39	..	..	122	..
Stockholm, Hornsgatan	April - June 97	36	..	..	113	..
Stockholm, Hornsgatan	April - June 98	40	..	..	149	..
Sundsvall, Skolhusallén	Winter 97–98	20	85	..	..	..
Norrköping	Feb - May 93	25	63	..	80	242
Umeå, Storgatan	Jan 94 - Jun 95	13	..	105	..	..
Sollentuna, Kanalvägen	Winter 95–96	36	93	97	106	259
Sollentuna, Kanalvägen	Summer 96	35	71	86	123	347
Sollentuna, Pommern- vägen	Summer 96	29	..	77	81	197
Sollentuna, Pommern- vägen	Winter 96–97	26	62	64	89	189
Sollentuna, Pommern- vägen	Summer 97	21	45	45	55	180
Sollentuna, Pommern- vägen	Winter 97–98	24	69	90	84	249
Sollentuna, Pommern- vägen	Summer 98	25	78	95	89	189

of Sweden. PM_{10} and $PM_{2.5}$ were studied in Lycksele, a small city in the inland area of northern Sweden and in Göteborg during 1 month in March–April 1997 (37). Similar concentrations were recorded at the two sites; the average PM_{10} concentration was 26 and 28 $\mu\text{g}/\text{m}^3$ in Lycksele and Göteborg, respectively, the average $PM_{2.5}$ concentration being 13 and 15 $\mu\text{g}/\text{m}^3$, respectively.

In summary, the concentrations of particles in the cities and municipalities in northern Sweden may be about the same as in the southern part. The influence of long-range transport is strong in the south, but decreases with latitude, while the importance of low dispersion during inversions increase with latitude.

Comparison with guidelines and limit values

There is no limit value for the concentration of PM_{10} in urban air in Sweden. However, there are guidelines stating that the 98th percentile of the 24-hour mean should be less than 110 $\mu\text{g}/\text{m}^3$ and the mean of the winter season (1 October — 31 March) should be less than 50 $\mu\text{g}/\text{m}^3$. All reported data are below the guideline levels. Limit values for PM_{10} have recently been introduced in the European Union. In the first step, to be reached before 2005, the 90th percentile of the 24-hour means should be less than 50 $\mu\text{g}/\text{m}^3$ and the annual mean less

than 40 $\mu\text{g}/\text{m}^3$. In the planned second step, to be reached in 2010, the 98th percentile of 24-hour means should be less than 50 $\mu\text{g}/\text{m}^3$ and the annual mean less than 20 $\mu\text{g}/\text{m}^3$. It is evident that the present concentrations at street levels in Swedish cities (see the preceding text) are higher than the limit values that are planned for introduction in 2010. However, the second step may be changed due to a revision that will take place in 2003.

Rural background PM_{10} concentrations

As already noted, the rural background concentration of PM_{10} in the Stockholm region is about 10 $\mu\text{g}/\text{m}^3$. A summary of the PM_{10} results obtained at a rural background site is shown in table 10. There are no data from other parts of the country. However, the concentrations of PM_{15} and $PM_{2.5}$ at a rural site in northern Finland during 1992–1994 was 5.3 $\mu\text{g}/\text{m}^3$ and 3.7 $\mu\text{g}/\text{m}^3$, respectively (39). As $PM_{2.5}$ made up almost 80% of the PM_{15} and as the conditions in northern Finland are similar to the ones in northern Sweden, the PM_{10} concentration in northern Sweden is estimated to be 4–5 $\mu\text{g}/\text{m}^3$. In Aspvreten, at the rural station 100 km from Stockholm, the concentration of particulate sulfate was about 75% of the levels found in Vavihill, the location of a background site near Malmö. On the assumption that the same relation is valid for PM_{10} , the background concentrations of PM_{10} in the far south of Sweden can be estimated to be 13–14 $\mu\text{g}/\text{m}^3$.

Seasonal variation

The seasonal variation (ie, the difference between the highest and lowest monthly means) in PM_{10} is rather small at roof level (figure 3). In Göteborg, the highest concentrations have been measured in February through May. In contrast, no seasonality is apparent at the rural site Aspvreten. The increased difference between the two stations in spring may be explained by the high contribution of resuspended road dust in the city in spring. The increased resuspension is due to the build-up of particles on the road surface during sanding and salting, as well as to road wear from studded tires in the winter season.

Comparison with PM_{10} concentrations in other countries

Different sampling methods and sampling site criteria make it difficult to compare PM_{10} data obtained in different parts of the world. Roof-level measurements are used in Sweden to estimate the urban background, while ground-level measurements located away from any direct sources are often used elsewhere. The TEOM sampler is totally dominant in Sweden at present, but gravimetric methods are used in other countries.

Table 10. Statistical summary of PM_{10} daily average concentrations at Aspvreten. (N = number of daily averages used in the calculations)

Year	Mean ($\mu\text{g}/\text{m}^3$)	Median ($\mu\text{g}/\text{m}^3$)	98th percentile	Maximum ($\mu\text{g}/\text{m}^3$)	N
1990	13.6	9.7	55.6	>100.0	219
1991	12.1	9.2	37.3	61.4	262
1992	12	9.2	33.6	54.6	229
1993	11.8	9.2	33.8	50.4	270
1994	10.4	8.9	28.0	33.9	321
1995	10.5	8.4	29.3	39.2	284
1996	9.5	7.2	33.2	48.1	318

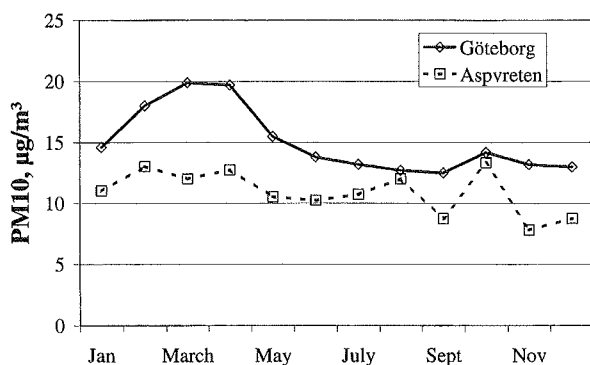


Figure 3. Seasonal variation of PM_{10} (particulate matter <10 μm in diameter) in a town, Göteborg, and at a rural site, Aspvreten, in southern Sweden — average 1990–1998.

Table 11. Summary of concentrations of PM₁₀ and PM_{2.5} in various parts of Europe and the United States. (90th percentile values = daily means).

Area	PM ₁₀		PM _{2.5}		Location	Study period	Number of sites	Method	Reference
	Mean or median	90th percentile	Mean or median	90th percentile					
The Netherlands	41	88	..	..	Urban background	Winter 1992—1993, Winter 1993—1994	1	Filter	40
The Netherlands	34	91	..	..	Nonurban	Winter 1992—1993, Winter 1993—1994	1	Filter	40
The Netherlands	29	51	14	38	Urban background	Winter 1994—1995	1	Filter	40
The Netherlands	24	44	15	34	Nonurban	Winter 1994—1995	1	Filter	40
Scandinavia	13—23	25—40	..	..	Urban background	Winter 1993—1994	4	Filter	17
Central Europe	40—75	80—150	..	..	Urban background	Winter 1993—1994	6	Filter	17
Scandinavia	11—16	20—30	..	..	Rural	Winter 1993—1994	4	Filter	17
Central Europe	40—75	60—100	..	..	Rural	Winter 1993—1994	6	Filter	17
United Kingdom	20—30	..	..	..	Urban background	1993—1994	16	TEOM	18
Canada	28	47	14	26	Urban background	1984—1993	11	Filter	41
Canada	11—30	..	7—17	..	Rural	1984—1993	4	Filter	41
Western United States	6—20	..	3—6	..	Rural	1988—1993	28	Filter	42
Eastern United States	10—20	..	8—14	..	Rural	1988—1993	11	Filter	42
Alaska	4	..	2	..	Rural	1988—1993	1	Filter	42
Northeast United States	17—29	..	10—15	..	Urban background	1988—1994	>100	Filter	5
New York, city center	50—60	70—100	20—30	50—80	Urban background	1988—1994	1	Filter	5
Sweden	10—20	..	..	..	Urban background	1990—1998	6	TEOM	This report
Sweden	11	22	8	14	Rural	1990—1998 ^a	1	TEOM	This report

^a PM_{2.5}: 1998.

Therefore, the reported PM₁₀ concentrations in Sweden tend to be lower than concentrations reported from other parts of Europe (table 11). In western Europe, the average daily concentrations are 20—35 µg/m³ in the urban background air. In central and eastern Europe, the average concentration in major urban areas is 40—75 µg/m³, the 90th percentiles of the daily means being 80—150 µg/m³. The difference between rural and urban background concentrations in central Europe averages less than 25% and thus illustrates the importance of long-range transport on the PM₁₀ levels. The urban concentrations in Canada and in humid areas of the United States are similar to the ones found in western Europe.

PM_{2.5} concentrations

Data on PM_{2.5} are very sparse. About 1 year of measurements in Stockholm and at a rural site 100 km south of Stockholm indicate that the PM_{2.5} fraction of PM₁₀ is 40—50% at street level and about 70—80% at roof level and in background air. These findings confirm results from studies in Canada, the United States, and The Netherlands (table 11).

Time trends of PM₁₀

The results given in table 9 do not indicate any time trend for PM₁₀ in the urban background (roof level) in

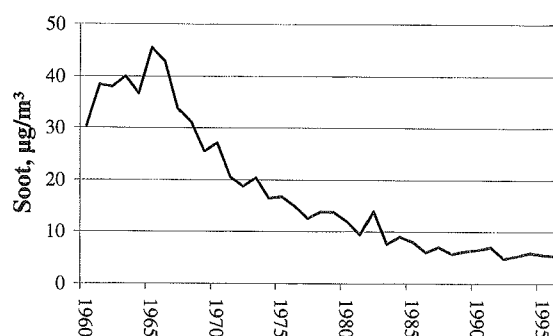


Figure 4. Concentrations of soot (black smoke) (annual mean, µg/m³) in central Göteborg in 1960—1996. Source: Göteborgs stads miljöförvaltning, 1997 (43).

the 1990s. Very few data are available before 1990. However, PM₁₀ concentrations in Göteborg have decreased from about 60 to about 15 µg/m³ since 1975 (43). The observed trend should be treated with caution, as the sampling method and site have changed during the period. The concentration of soot (BS), which can be seen as an indirect measurement of particles, has decreased substantially in Göteborg, from 45 to 5 µg/m³, since the beginning of the 1960s (figure 4). The remarkable decrease reflects the large efforts that have been made in Sweden to reduce the emission of several pollutants during the last few decades. For example, the Swedish emission of SO₂ in 1995 was 80% less than the

emission in 1980. Similar reductions have taken place in other parts of western Europe. As the ratio between soot and PM₁₀ concentrations is time and site dependent, the observed trend cannot be directly translated to PM₁₀. However, it is clear that the PM₁₀ levels have also decreased substantially.

As already noted, there are no trends for urban background. However, at the rural site in Aspöreten, the annual mean PM₁₀ concentrations decreased about 25% from 1990 to 1996 (table 10).

Future trends

Predicting the future trend for the concentrations of aerosols in the air depends on judgments and assumptions regarding future changes in the emissions of the sources that contribute most to the particle levels found in air today.

In Sweden, long-range transport is the predominant source of PM₁₀ in rural areas, as well as in the urban background, in southern Sweden. The contribution from locally emitted particles to the PM₁₀ concentration is estimated to be 3–7 µg/m³ in the urban background air. In street canyons, road traffic (direct emissions and re-suspension of road dust) add, on the average, another 10–30 µg/m³. However, the addition varies greatly, as especially the amount of re-suspended dust varies substantially. The highest concentrations are generally found in spring when re-suspension is greatest. Wood combustion for residential heating may be a very important source in certain areas, but no estimation of its contribution is available.

The future contribution from the long-range transport of particles depends on the trend of emissions in Europe. Tarrasón & Tsyro (35) have calculated the concentrations of inorganic PM_{2.5} particles in Europe for 2010, on the assumption that the emission of precursor gases in 2010 will be in accordance with the current European emission reduction plans. Although the particle concentrations will decrease by 25–30% in central Europe, the concentrations will be essentially unchanged in most of Sweden. The planned emission reductions in countries of western Europe will be counteracted by increased emissions in eastern Europe.

It has been estimated that the emissions of primary particles per vehicle from gasoline and diesel vehicles will decrease in the future due to the continuing introduction of cars equipped with catalysts and improved diesel engines. However, the reductions in PM emissions envisaged as a result of improved technology could be in danger if the percentage of diesel cars, which emit considerably more than gasoline cars, increases too much. The share of diesel-fueled cars among new cars in Sweden has increased from about 1% in 1990 to more than 10% at present (44).

The future emission of particles from the burning of wood for residential heating depends on several factors. If modern technology is introduced, the emission of PM will decrease substantially. As the life-time of boilers is normally 25–40 years, the shift from old to new technology is expected to be slow. An increase in the use of wood and other biomasses as fuel due to higher prices for oil and electricity will counteract the reduced emissions due to better combustion technology.

The emission of coarse particles due to the resuspension of road dust will probably not change in the future. The emissions due to studded tires may decrease if the use of new stud-free winter tires increases. Better and more frequent cleaning of the streets, particularly in spring, would also reduce the amount of dust available for resuspension. Less frequent sanding and salting would produce the same effect, although it might have other unwanted consequences.

In summary, emission trends are difficult to predict for the future, as they depend on several factors. However, it seems unlikely that there will be any dramatic changes during the next 10–15 years.

Human exposure

When relating human health effects to exposure to PM (as in epidemiologic studies) time-weighted-average ambient concentrations are often used as a surrogate for personal exposure because, under normal circumstances, only fragmentary data are available on true personal exposures. Further extrapolation to the dose is impossible in practice, as individual ventilation rates and the pulmonary deposition rates are affected by unmeasured individual physiological parameters. Moreover, the deposition is dependent on the normally unknown particle-size distribution in the air actually inhaled by each person.

The personal exposure (ie, the total PM exposure of a person during a specific time period) is composed of exposure to many different particles from various sources in different microenvironments. A microenvironment can be defined as a volume in space, during a specific time interval, during which the variance of concentration within the volume is significantly less than the variance between that microenvironment and its surrounding microenvironments (45). Microenvironments can be of very different sizes in both space and time. They are normally small in both time and space near a source (eg, a smoker) and increase in size spatially and temporally in the absence of any nearby sources.

A person can pass through a large number of microenvironments during a day (normally the shortest time period used in epidemiologic studies); however, differ-

ent microenvironments can be divided into the following four general classes: indoor, ambient-outdoor, occupational, and in-traffic. The relation between indoor and ambient-outdoor concentrations of particles is especially important as people in industrialized countries spend most of their time indoors. A time budget survey in the United States showed that US residents spend 87.2% of their time indoors (mainly at home or at work), 7.2% in other nonambient environments (eg, in vehicles in transit), and only 5.6% outdoors (46). A time-budget examination in Sweden, including 160 children in the age range of 7–12 years, gave similar results. The children spent an average of 9% of their time outdoors (47). However, as the amount of physical activity outdoors is usually larger than that indoors, the ventilation rate, and thus the dose (the amount of deposited particles in the respiratory tract), is proportionally more important than the time spent outdoors.

The US EPA (5) has evaluated several mainly US studies in determining the relative contributions of ambient-outdoor and nonambient (indoor) particles to total human exposures. The key conclusions were as follows:

- The ambient environment can be a major source of indoor particles due to air exchange and infiltration. Whether it is the dominant source depends on the relative magnitude of the indoor sources of particles.
- The largest identified indoor source of particles in both homes and other buildings is cigarette smoke. The $PM_{2.5}$ concentration in homes with smokers has a 24-hour average increment ranging between 25 and 45 $\mu g/m^3$. The second largest identified source is cooking. In homes with cooking, the PM_{10} 24-hour average concentrations increase by 10 to 20 $\mu g/m^3$.
- Indoor air concentrations of PM_{10} and $PM_{2.5}$ generally exceed outdoor concentrations except in areas where outdoor concentrations are high.
- The regression of daily indoor PM_{10} and $PM_{2.5}$ concentrations on the corresponding daily outdoor concentrations generally explains less than one-half of the variance ($r^2 < 0.5$) if the regressions are carried out for all homes in the study. However, the regression for a single home (in cases in which homes were measured repeatedly) often has a better indoor-outdoor relationship (r^2 up to 0.9). Since many epidemiologic studies deal with repeated measurements over time, "longitudinal" regressions by individual homes may be more relevant to these studies than "cross-sectional" regressions across all homes.
- For PM, studies have detected a "personal cloud" related to the activities of a person who may generate significant levels of particles in his or her vicinity. The

personal exposure is thus often higher than the indoor and ambient concentrations.

- When a cross-sectional analysis is performed that compares ambient PM_{10} with individual personal exposures for a group of subjects, the correlation often goes towards zero, because of the large influence of indoor sources and sinks that vary *between* individuals. However, because of the relative consistency *within* any given residence for the indoor sources and sinks of PM_{10} , the longitudinal (time-series) correlation of personal exposure of a specific person to the total indoor PM_{10} (from outdoor *and* indoor sources) and ambient PM_{10} can be very high. Two analyses involving a limited number of subjects yielded correlation coefficients (R^2) above 0.9 when indoor sources were consistent from day-to-day.
- Both fine and coarse particles enter indoors. However, because coarse particles are rapidly removed by deposition, the indoor concentration of ambient fine particles is more representative of the outdoor, ambient, fine particle levels. Thus, in turn, a centrally located ambient monitor may provide a fine-particle concentration that is representative of community exposure (indoor and outdoor) to ambient fine particles, but not a representative sample for coarse particles.
- Indoor sources do not generate fine-mode particles of the same composition as the most common particles of outdoor origin (eg, sulfates, metals, etc). Moreover, the production of indoor-generated particles is controlled by daily indoor activities. Therefore, the exposure to indoor-generated particles will not correlate with the concentration of ambient (outdoor-generated particles), and time-series epidemiology based on ambient measurements will not identify health effects of indoor-generated particles.
- Therefore, the measurement of *daily* variation of *ambient* concentrations of PM, as used in time-series epidemiology studies, has a plausible link to the daily variations of human exposure to particles from ambient sources, for the people represented by the ambient monitoring stations. This link should be better for indicators of fine particles ($PM_{2.5}$) than for indicators of fine plus coarse particles (PM_{10} or TSP), which in turn should be better than indicators for coarse particles ($PM_{10} - PM_{2.5}$).

Ultrafine particles

Recent work has suggested that very small particles (less than about 0.1 μm) may be responsible for some of the health effects associated with exposure to PM. (See the

section on ultrafine particles in this volume.) The hypothesis for explaining the health effects of these small particles is based on the number, composition, and size of the particles rather than on the mass, as they contribute very little to, for example, the PM_{10} concentrations.

The number of particles is a good measure of the load of these particles as the number concentration in ambient air is dominated by particles <0.1 μm . Particle concentrations vary widely from $100/cm^3$ in marine aerosol and around $1000/cm^3$ in clean continental backgrounds to over $100\,000/cm^3$ in polluted urban aerosols. The average residence time of these particles is strongly dependent on the number concentration as the probability to interact (coagulate, condense) with other particles increases when the number concentration increases. Thus the less the number concentration, the longer the residence time. The residence time of 10 and 100 nm particles is 1 and 10 days, respectively, in clean air ($300/cm^3$), but 15 minutes and 1 day, respectively, in polluted air ($15\,000/cm^3$).

Due to the short residence time of small particles, the number concentration is high close to sources and decreases rapidly with distance from the source. The number concentration near sources varies widely in time due to fluctuations in meteorology and source strength. Due to the short residence time, the number geometric mean diameters of the size distribution of aerosols increases from 10 nm in "freshly" polluted urban air to about 80 nm in "aged" air, found in clean background regions.

Summary and concluding remarks

Airborne PM is a very diverse material in terms of its physical and chemical properties, and many sources contribute to the atmospheric concentrations. Aerosol particles are typically very small, ranging in size from a few nanometers up to ≥ 100 μm . While all particles in this range can be found in the air, only particles in the range of 0.1–2 μm can remain airborne for long periods (≥ 10 days in clean air).

Particles less than about 10 μm (termed PM_{10}) are the most important from a health point of view, as larger particles do not reach the thoracic parts of the lung. In recent years, particles <2.5 μm (termed $PM_{2.5}$) have been suggested as more relevant for studies of the health effects of airborne particles.

Airborne particles may be of either primary or secondary origin. Primary particles, formed by mechanical processes or in combustion processes, are directly emitted to the air. Secondary particles are formed in the air by chemical reactions or the condensation of gases. Me-

chanically formed particles are normally larger than 2.5 μm exposure (termed coarse particles), while the particles formed from gas conversions are smaller than 2.5 μm (termed fine particles). Due to their different size and formation processes, fine and coarse particles have substantially different properties (chemical composition, atmospheric life-time, temporal and spatial variability).

The annual Swedish anthropogenic emission of primary particles is estimated to be 42 000–100 000 t. Residential wood burning for heating is probably the major source, but the emission estimate is based on limited data. Heavy-duty diesel vehicles, the combustion of heavy oil for energy production, machinery, and industrial processes are other important sources. The resuspension of road dust is probably another important source, but no reliable estimates of its contribution are available on the national level. Industrial emissions and the resuspended dust consist mainly of coarse particles; all other sources emit mainly fine particles. The formation of secondary particles due to emissions from various sources is not included in the emission estimates.

The rural background PM_{10} concentration in the Stockholm region (south central Sweden) is about 10 $\mu g/m^3$. The background concentration in the far north can be estimated to be 4–5 $\mu g/m^3$ based on data from northern Finland. PM_{10} concentrations in the far south can be estimated to be 13–14 $\mu g/m^3$, if the ratio between particulate sulfate concentrations in central and south Sweden is also assumed to be valid for PM_{10} .

Regular monitoring of PM_{10} in a few major cities in southern Sweden since the beginning of the 1990s shows that the average urban background concentrations are 12–20 $\mu g/m^3$. The narrow range is due to the strong influence of the long-range transport of particles on the concentrations in urban backgrounds. The 98th percentiles of the daily mean values range from 30 to 50 $\mu g/m^3$, the maximum daily mean concentrations are in the range of 50–100 $\mu g/m^3$. At street level, the mean concentrations range from 20 to 40 $\mu g/m^3$, the 98th percentiles of the daily means being 50–100 $\mu g/m^3$. The maximum daily concentrations range from 100 to 150 $\mu g/m^3$, and hourly mean concentrations can reach 300 $\mu g/m^3$. Data on PM_{10} in the towns and municipalities of northern Sweden are sparse. However, regular BS measurements at a number of sites in the north and short-term PM_{10} measurements in a typical inland municipality show that the concentrations in the cities and municipalities in northern Sweden may be about the same as in the southern part. The influence of long-range transport is strong in the south, but decreases with latitude, while the importance of low dispersion during inversions increases with latitude.

The seasonal variation (ie, the difference between the highest and lowest monthly means of PM_{10}) is 5–10 $\mu g/m^3$ in the urban background. PM_{10} concentra-

tions are highest in spring, possibly because of the high contribution of road dust resuspension due to the build up of particles on the road surface during sanding and salting, as well as road wear from studded tires in the winter season.

Urban background PM₁₀ concentrations in Sweden are generally lower than in other parts of Europe. In western Europe, the concentrations average 30% to 130% higher than the ones found in Swedish cities. In central and eastern Europe, the average concentration in major urban areas is 2.5 to 5 times the Swedish urban background. However, different sampling methods and sampling site criteria may introduce bias into the comparisons.

There was no time trend for PM₁₀ in Swedish cities in the 1990s. Very few data are available before 1990, making longer trend analyses very uncertain. However, the large reduction of particle precursors in Sweden, as well as elsewhere in Europe, justifies the assumption that the particle concentrations have decreased. The assumption is supported by the observation that the PM₁₀ concentrations in Göteborg have decreased from about 60 to about 15 µg/m³ since 1975. The magnitude of the observed trend should be treated with caution, as the sampling method and site have changed during this period. The assumption is also supported by the large decrease of BS, an indirect measurement of particles, from 45 to 5 µg/m³, since the beginning of the 1960s in Göteborg.

No major changes of the concentrations are anticipated during the next few decades. There are indications that reduced emissions due to better combustion technology for vehicles and wood boilers will be counteracted by an increased share of diesel cars and an increased use of wood for heating. Moreover, the contribution from long-range transported inorganic fine particles has been estimated to be about the same in 2010 as now.

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