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AEROSOLS: CONNECTION BETWEEN REGIONAL CLIMATIC CHANGE AND AIR QUALITY

(IUPAC Technical Report)

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Aerosols: Connection between regional climatic change and air quality

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Abstract: Aerosols play an important role in all problems connected with air pollution, ranging from very local effects and human health problems to regional problems such as acid deposition and eutrophication up to continental and global questions such as stratospheric ozone loss and climatic change. In this report, an explanation of these effects is given and an assessment is made for parts of China, based on the aerosol data given by Zhang et al. elsewhere in this volume.

Epidemiological research has made clear that aerosols are a cause of enhanced mortality due to cardiopulmonary diseases (heart and lung diseases). Based on the same mortality as found in Europe and on linear extrapolation (with large uncertainties; no sufficient data are available to make better estimates), an excess mortality of 5000 to 10 000 due to acute effects and 20 000 to 50 000 due to chronic effects per year could be expected for a city like Beijing with a population of 14.5 million. A major cause of these uncertainties is problems in the determination of semivolatile compounds and elemental or black carbon in aerosols.

Aerosols have a strong impact on the radiative balance of the earth, in a direct way by reflecting solar light as well as in an indirect way by cloud formation leading to clouds with higher albedo, which reflect sunlight better. The total direct effect, backscatter of sunlight, including backscatter from nitrates and organic compounds, is estimated to be approximately 2 to 3 W m^{-2} for Western Europe, while the indirect effect is approximately 0 to –6 W m^{-2}. Soot absorbs incoming solar radiation and heats the atmosphere. This process contributes 0.1 to 0.2 W m^{-2} on a global scale.

If PM-2.5 levels are compared with Europe, a direct effect of approximately –4 to –10 W m^{-2} would be plausible for China, black carbon could contribute probably about 0.5 to 2 W m^{-2}, and the indirect effect could be about 0 to –6 W m^{-2}. These effects could cause a net cooling over China (and over many developing countries in the same position) of about –4 to –15 W m^{-2}. This estimate is obviously based on many assumptions and hence is quite uncertain.

One must be aware that measures reducing local aerosol concentrations will have a large impact on the radiative balance and could, over a few decades, have potentially at least the same impact as the build-up of greenhouse gases since the beginning of the Industrial Revolution.

1. THE PROBLEM

Aerosols, defined as small particles suspended in air with a lifetime of at least some minutes, are either emitted as such (primary aerosols, dust, or as particle emissions by diesel-powered cars) or they are formed by the conversion of sulfur dioxide, nitrogen oxides, ammonia or organic compounds in atmospheric chemical reactions to sulfates, nitrates, ammonium compounds, or nonvolatile organics (secondary aerosols).

Particles with an aerodynamic diameter below 0.05 µm disappear rapidly because they diffuse to other particles and are lost by conglomeration. Particles over 15 µm in aerodynamic diameter are deposited relatively rapidly by gravitational settling. Particles between 0.1 and 2 µm in aerodynamic diameter live quite long, up to 1 or 2 weeks, as their main sink is to act as nuclei for the formation of cloud droplets. An overview of the number concentration, mass concentration, and lifetime for aerosols as a function of size in polluted areas is given in Fig. 1.

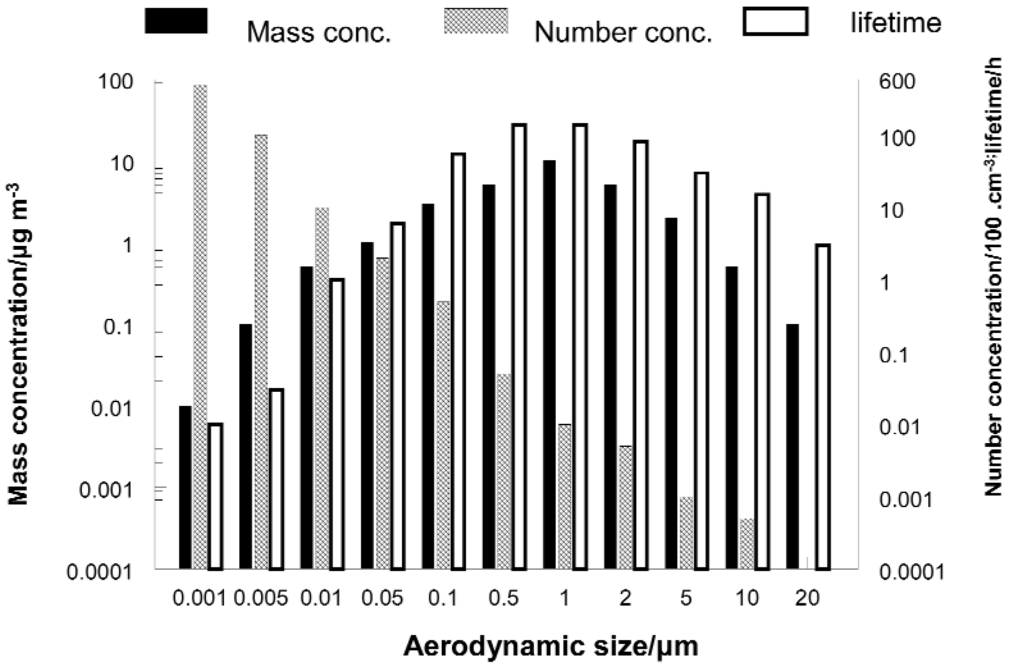


Fig. 1 Lifetime, number concentration (right axis) and mass (left axis). The distribution is typical for a roadside in a Dutch city. Further away from traffic, the contribution of particles with aerodynamic diameters smaller than 0.05 µm is much smaller.

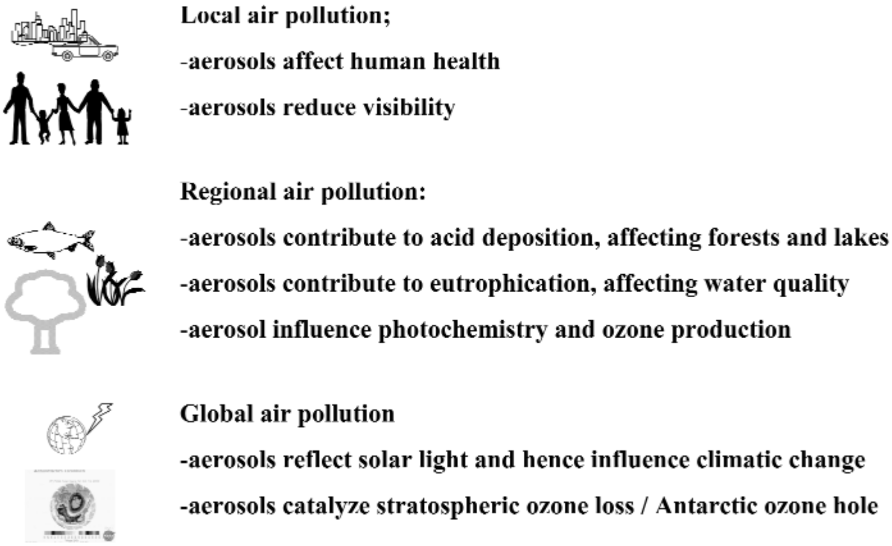
The large number of particles with aerodynamic diameter below 0.1 µm is caused by traffic emissions; all Otto and Diesel engines cause huge emissions of very small particles with very small mass and, as they coagulate rapidly, short lifetimes. Aerosol particles between 0.2 and 2 µm comprise a large fraction of the mass; their numbers are much smaller than the small particles, but they live a relatively long time, so most mass is gathered in this size range. Large particles are characterized by low number, large mass, and short lifetimes; they fall out of the atmosphere, so to speak.

It has become clear that aerosols play an important role in most air pollution problems, ranging from very local effects and human health problems to regional problems such as acid deposition and eutrophication to continental and global problems such as stratospheric ozone loss and climatic change. An overview of all the aerosol-related air pollution problems is given in Fig. 2.

- Epidemiological research has made clear that aerosols have a large impact on human health. Aerosols are not only the cause of enhanced mortality due to cardiopulmonary diseases (heart and lung diseases), but also contribute to respiratory illnesses such as asthma and bronchitis.
- Deposition of ammonium, sulfate, and nitrate in aerosols constitutes an important contribution to acid deposition and eutrophication (damage to ecosystems due to large input of nutrients such as nitrate), and can be 30 % or more in Western Europe.
- Heterogeneous reactions at the surface of aerosols contribute (probably) very significantly to ozone formation in the troposphere while reactions at the surface of aerosols in the stratosphere are a main factor in the destruction of stratospheric ozone by CFCs (chlorofluorocarbons).

- Aerosols have a large impact on the radiative balance of the earth and hence on climatic change by reflecting incoming sunlight, by cloud formation, or by enhanced absorption of light by soot.

This complex situation entails that any measures that influence the concentration of ambient aerosols will have an impact on many issues simultaneously. For instance, abatement policy to reduce the impact of aerosols on health, a very local issue, will have consequences for the regional radiative balance and hence on regional climatic change. Local air quality and regional climatic change are connected by a common factor: aerosols.



Local air pollution;
-aerosols affect human health
-aerosols reduce visibility

Regional air pollution:
-aerosols contribute to acid deposition, affecting forests and lakes
-aerosols contribute to eutrophication, affecting water quality
-aerosol influence photochemistry and ozone production

Global air pollution
-aerosols reflect solar light and hence influence climatic change
-aerosols catalyze stratospheric ozone loss / Antarctic ozone hole

Fig. 2 Overview of all aerosol-related air pollution problems.

2. IMPACT OF AEROSOLS ON HUMAN HEALTH

Many attempts have been made in the past to trace the impact of air pollution on human health, but to no avail. Developments in epidemiological methodology as well as better monitoring methods have changed that situation, and recent epidemiological research has made clear that fine particular matter (aerosol particles smaller than 10 µm in aerodynamic diameter) have an impact on human health [1–4].

The principle of the time series method is based on comparing the actual mortality with the mortality calculated by way of the known impact of different diseases, accidents, etc. A small fraction of typically a few percent is left which cannot be explained this way. This fraction correlated with different possible factors, and a good correlation with air pollution is found. More detailed investigations show (at least for Europe and the United States) that no correlation is found with, e.g., sulfur dioxide concentrations, but a statistical significant correlation is found with the concentration of fine particles, PM-10 or PM-2.5.

This effect of aerosols on human health has been traced for different locations, but the excess mortality varies. The effect seems to be linear, and no effect threshold has been found [4].

Table 1 Results of epidemiological investigations of excess mortality caused by aerosols [4].

Location	Excess mortality
USA	2–4 % for an increase of ambient aerosol of 100 μg m^{-3}
Western Europe	2 % for an increase of ambient aerosol of 100 μg m^{-3}
Eastern Europe	1–2 % for an increase of ambient aerosol of 100 μg m^{-3}
Western Europe	3 % for an increase of "black smoke" of 50 μg m^{-3}
Eastern Europe	0.6 % for increase of "black smoke" of 50 μg m^{-3}

Time series epidemiological research indicates that the increased death rate is 1–2% for the Netherlands. This means, at the Dutch ambient PM-10 level of 32 μg m^{-3}, an excess acute death rate of 1700 to 3000 death per year due to aerosols, on a population of 16 million.

It is much more difficult to assess the chronic effects of exposure to aerosols, and the estimates of these chronic effects are quite uncertain, but could be about 10 000 to 15 000 for the Netherlands [4].

The other epidemiological method used to trace the impact of aerosols is the cohort method. In the cohort method, a large group of people is characterized by dividing according to those parameters which are under investigation, so groups can be formed, e.g., smoker/nonsmoker, living near road/living further away, etc. Mortality and frequency of diseases are estimated next as a function of these parameters, and the different groups are compared. Statistical techniques can "tease apart" different parameters.

Cohort studies are needed to quantify the shortening of life by aerosols (which in the case of a large Dutch cohort study is estimated to be 1 to 2 years) but also to discriminate between different factors [5]. In this large Dutch cohort study, groups were sorted as a function of living near or further away from roads with a lot of traffic. Traffic emissions generate clearly large gradients of aerosol number concentrations, the extra mass contribution to PM-2.5 or PM-10, but also of soot (indicated as EC, elemental carbon) as a function of the distance from roads is limited; see Figs. 3a and 3b.

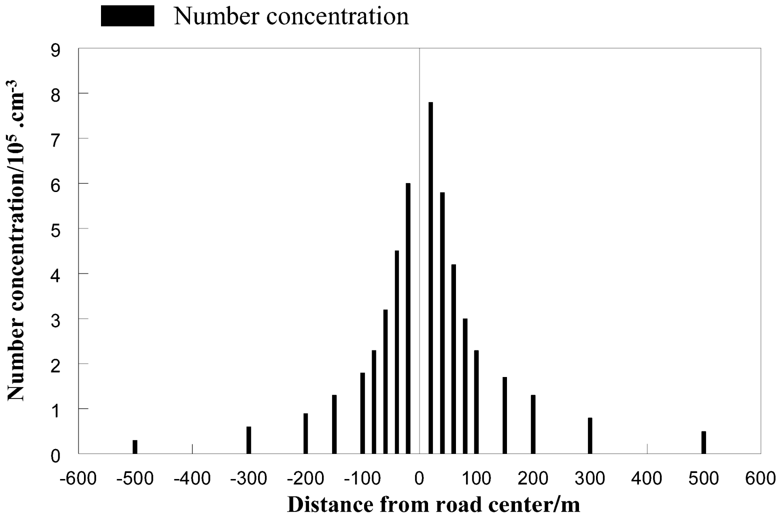


Fig. 3a Particle number concentration as function of distance to a busy road in the Netherlands.

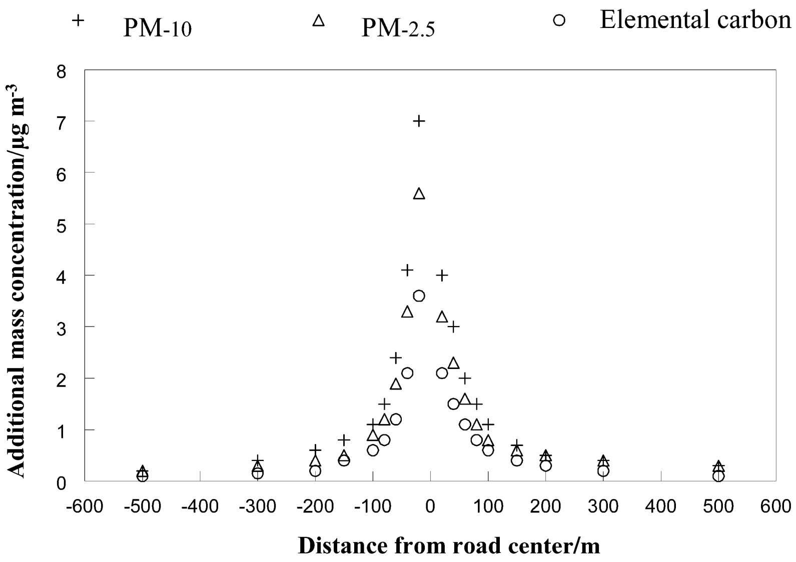


Fig. 3b Modeled gradients of PM-10, PM-2.5, and elemental carbon due to road emissions at a busy road in Rotterdam, Netherlands, based on measurements in 2000/2001.

The results of this cohort study indicate that excess mortality due to cardiopulmonary diseases is twice near roads compared to locations further away from roads.

Neither toxicological nor epidemiological research has yet revealed which fraction of aerosol, PM-10 (mass of aerosol particles with an aerodynamic diameter <10 µm), PM-2.5 (mass of aerosol with an aerodynamic diameter <2.5 µm) or ultrafine particles or specific compounds in aerosol particles are responsible for the observed health effects. The results of this Dutch cohort study indicate that PM-2.5 or ultrafine particles (aerosol with an aerodynamic diameter <0.1 µm) could have a large impact.

It is clear that excess mortality due to aerosols in, e.g., Chinese cities could be quite high. Zhang et al. report elsewhere in this volume that the PM-10 concentrations in large Chinese cities can be between 50 and 120 µg m^{-3}. If the same excess mortality is assumed as found in Europe and linear extrapolation is made (which are both quite uncertain assumptions; insufficient data are available to make better estimates), then an excess mortality of 5000 to 10 000 due to acute effects and 20 000 to 50 000 due to chronic effects per year could be expected for a city like Beijing, with a population of 14.5 million. Apart from this excess mortality, an excess number of cases of asthma and bronchitis as well as heart diseases can be predicted.

It is obvious that such an impact of aerosols on human health will result in a large economic loss, so air quality standards are in place in Europe (yearly average PM-10 of 40 µg m^{-3}) or are under consideration (USA: PM-2.5 standard of 15 µg m^{-3}) and measures are taken to meet these standards.

No mechanism has been found to explain the effects of aerosols, and correct measurement of the mass and chemical composition of aerosols pose many problems, which impose severe hindrances in formulating and imposing effective air quality standards for aerosols.

3. PROBLEMS IN THE CHARACTERIZATION OF PARTICULATE MATTER

Filter-based methods that are in general use need sampling times of 5–24 h, rather long for health-related studies. Even more important, they lead to underestimation of volatile components, as excess sulfate can be produced from artifacts caused by topological reactions, they are plagued by high and variable blanks and, last but not least, are labor-intensive.

The loss of semivolatile components has been known for a long time and has been documented in detail in the literature [6–8]. Evaporation of compounds such as ammonium nitrate and semivolatile organic compounds is in most cases the most important source of errors and its implications for health studies and on the radiative forcing of aerosols are now generally perceived as important [5]. Investigations into these artifacts have led to the following general conclusions. As long as the sampled gases and aerosols are in equilibrium with each other and with the aerosols already sampled on the filter, artifacts are generally avoided. This means that if sampling periods are very short, a few hours or less, the results are much better compared to those obtained with long sampling periods (24 h or more)

Some filter materials, such as cellulose acetate and cellulose matrix filters, retain nitric acid quantitatively and also ammonia, at least if the concentration is not too high.

Losses of ammonium nitrate from filters of about 70 % have been found by Zhang et al. (see elsewhere in this issue) and their results are given in Fig. 4.

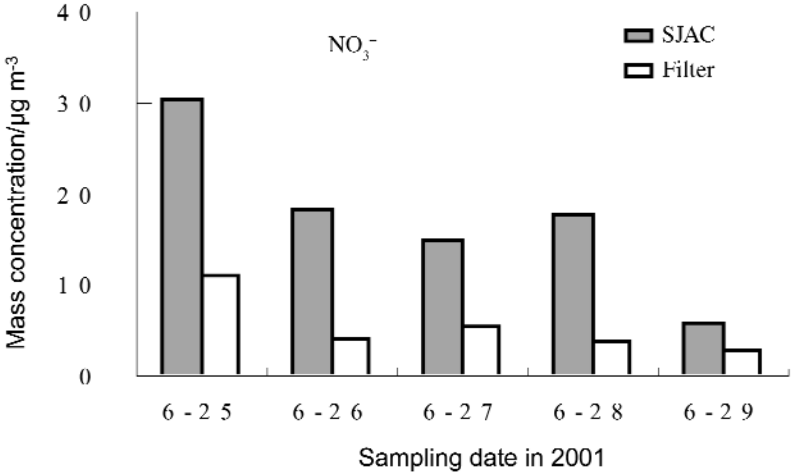


Fig. 4 Intercomparison between results obtained by filter methods and steam jet aerosol collector on a daily basis.

The steam jet aerosol collector (Fig. 5) is based on scavenging aerosol by mixing air with a steam jet and droplet formation [9].

A wet denuder system is used to scavenge interfering gaseous nitrogen compounds, in this case, ammonia and trioxonitric acid (nitric acid) and deoxonitric acid (nitrous acid). The absorption solution retains all gaseous interferences such as HNO_3, HNO_2, SO_2, and NH_3. This air stream is rapidly mixed with steam inside the mixing reservoir. The resulting high supersaturation causes aerosol particles to grow rapidly (within 0.1 s) into droplets of at least 2 µm aerodynamic diameter. These droplets containing dissolved aerosol species are then collected by a cyclone with a cut-off of 2 µm. The solution collected in the cyclone can be analyzed on- or off-line.

Not only does the measurement of major compounds in aerosol-like ammonium nitrate give severe problems, but also measuring the total PM-2.5 or PM-10 is problematic.

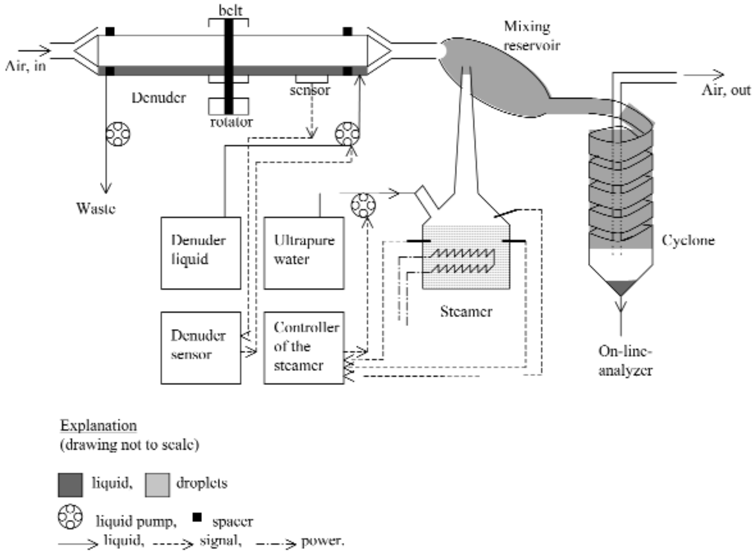


Fig. 5 Steam jet aerosol collector.

Two automated instruments have become quite popular the last years: the beta attenuation monitor and the TEOM (tapered element oscillating microbalance) [10]. An overview of the TEOM is given in Fig. 6. Particulate matter is sampled by way of a filter inserted in a tapered glass tube. The oscillating frequency of this glass tube depends on the mass on the filter, so the change of frequency is monitored and translated into the concentration of particulates in air. The filter has to be exchanged after 20 days, otherwise, the continual loading of the filter will reduce the accuracy of the measurement.

As water in aerosol is also recorded as mass, the sampled air is heated to remove water. The problem then is that semivolatiles (ammonium nitrate and certain classes of organics) are also removed.

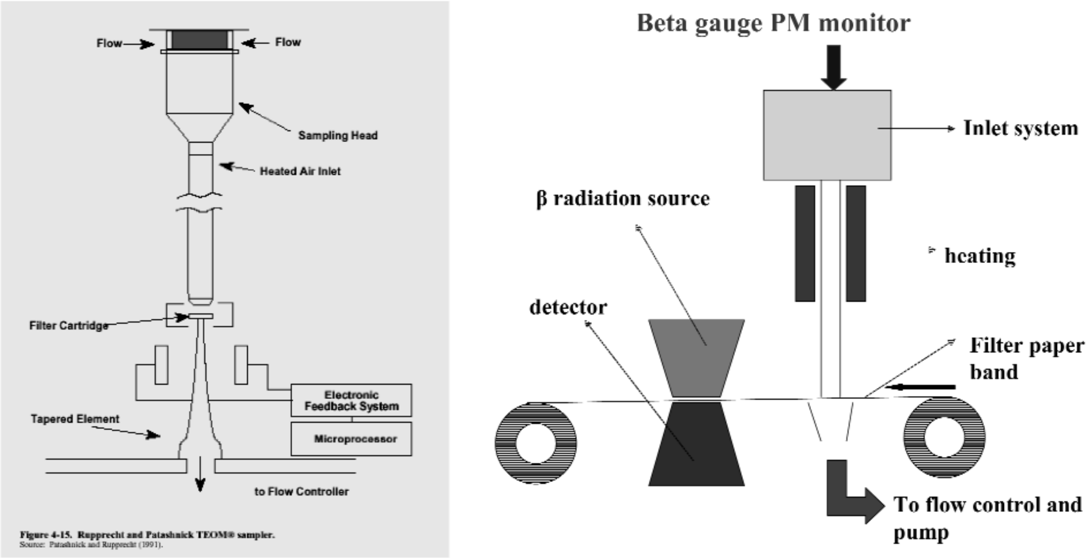


Fig. 6 Diagram of TEOM, left, and beta attenuation monitor, right.

The principle of the beta attenuator method involves sampling of particulate matter on a filter band and then transporting the band to the measurement location. Beta radiation consists of electrons, which have a limited ability to penetrate through matter. The penetration is a function of the characteristics of the filter band (which are constant) and the mass of sampled particulate matter. The method has the same drawback as the TEOM: to get rid of water in aerosol particles, the sampled air is heated and semivolatile compounds are lost.

Thus, for both methods a correction for these losses of 1.3 to as high as 1.9 has to be applied in some locations in Europe. This correction is not a constant as the amount of ammonium nitrate and organic compounds varies greatly, depending on place and time, even in a country as small as the Netherlands. It is obvious that these uncertainties in the characterization of aerosols introduce major problems in the interpretation of epidemiological results, calculation of the acid deposition flux, and the direct and indirect effect of aerosols.

The results reported by Zhang et al. (elsewhere in this volume), that a large fraction of ammonium nitrate in China is not sampled by filter methods, are supported by experiences in Europe: the total mass of PM-2.5 and PM-10 as well as the concentration of semivolatiles such as ammonium nitrate and semivolatile organics is probably underestimated due to the artifacts of currently employed measurement methodologies. The situation in developing countries could be worse, as indicated on the basis of the available data.

4. IMPACT OF AEROSOLS ON THE REGIONAL RADIATIVE BALANCE

The greenhouse effect is basically caused by absorption of long-wave radiation, emitted by the earth's surface, by greenhouse gases in the atmosphere. Solar radiation warms the surface. In order to reach equilibrium, the earth surface emits long-wave radiation. Part of this radiation is absorbed in the atmosphere, where CO_2, CH_4, N_2O, O_3, CFCs contribute significantly to the greenhouse effect. Most greenhouse gases live 10 years or longer, with the exception of ozone and water vapor, and are hence more or less uniformly mixed in the atmosphere. The shift in the radiation balance of these gases is of a global character.

Figure 7 shows the contribution on a global scale of the greenhouse gases, which contribute in total (including ozone) about 3 $W\ m^{-2}$ to the radiation balance, according to the International Panel on Climatic Change [11]. A small cooling effect by reflection of sulfate aerosol and a much larger indirect effect are indicated. Furthermore, some heating by black carbon (soot, elemental carbon) is also indicated.

"Aerosol" particles in the atmosphere reflect the radiation depending on their size distribution. The size of the particles determines whether radiation of shorter wavelengths is reflected more effectively compared to infrared radiation. If the wavelength of the incoming light and the size of the particle are about the same, backscatter will take place, i.e., the light is reflected back. Incoming solar radiation (where most energy is present between wavelengths of 0.4 to 1 μm) is effectively reflected by particles in the size range of 0.1 to 2 μm. Particles of this size do not intercept the outgoing infrared radiation of the earth [12].

In order to assess the cooling influence of this reflection, both the size and the chemical composition of aerosols must be known. The chemical composition influences not only scattering, but also determines whether a particle will grow by the uptake of water (vapor) due to the influence of the hygroscopic compounds of the aerosol. Smaller particles, with an aerodynamic diameter below 0.1 μm, are more abundant in the atmosphere compared to larger ones; see Fig. 1.

Until now, IPCC has emphasized the contribution of sulfate in the assessment of the radiative forcing of aerosols. It has been assumed in the past that nitrate is primarily present in particles with an aerodynamic diameter over 2 μm and hence would not make a contribution to radiative forcing. Measurements in the Netherlands, using the best available impactor technique with very short sampling

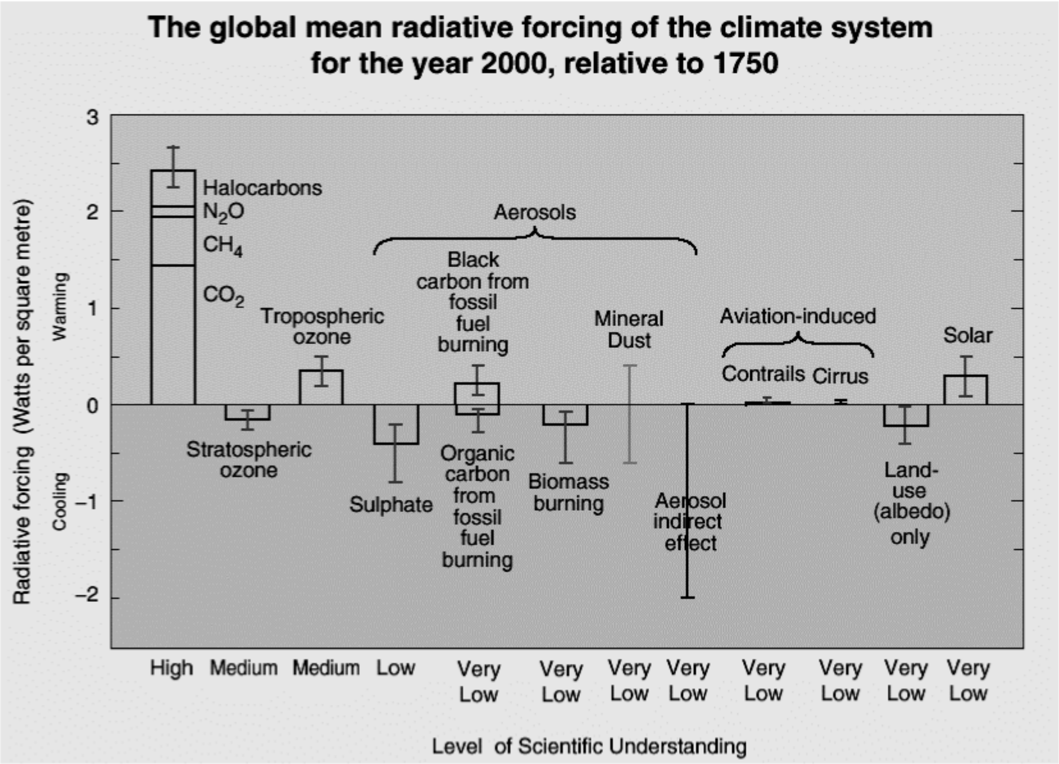


Fig. 7 Radiative balance according to the International Panel on Climatic Change (IPCC) [11].

times (30 min) to minimize losses, have shown that in Western Europe the same size distribution for nitrate as for sulfate is found [13,14].

The main problem is that only a little reliable information is available about the size distribution of nitrate in aerosols, even in developed countries. The high nitrate concentrations and the size distributions are illustrated in Fig. 8. These results were obtained by a combination of a Moudi impactor and an SJAC sampler.

The large contribution of nitrate is evident. The contribution of nitrate in the Netherlands is larger than sulfate [4].

Polar organic compounds were simply overlooked until recently. The oxidation of volatile organic compounds, either emitted by anthropogenic sources or biogenic, leads not only to the formation of ozone and other oxidants, but also to carboxylic acids, which are fairly polar compounds and water-soluble. These organic acids, and especially their salts, have very low vapor pressures and will hence be present in aerosols. Initial results of measurements both in the United States and Europe have already indicated that about 25 to 40 % of the aerosols consist of organic compounds. It has become clear that a large fraction of this organic material is made up of polar organic acids and related compounds. This material has the same size distribution as sulfates and nitrates and will contribute to the direct effect and indirect effect of aerosols in the same way as the inorganic compounds. The organic fraction in developing countries is quite high (see Table 2), and it could very well be that the impact of this organic fraction is as large or larger than sulfate. Information on the detailed composition, whether the compounds are soluble, etc., would be needed for a better assessment.

To make a more correct calculation, the composition of aerosol and others factors are needed. These data are not available for China, but if PM-2.5 levels are compared with Europe, a direct effect of about –4 to –10 W m^{-2} would be plausible for China; see Table 2.

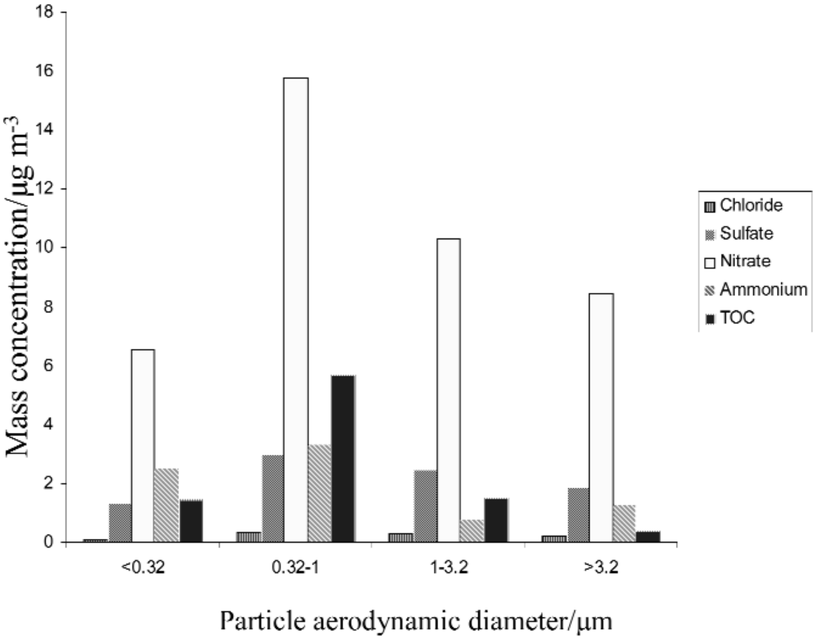


Fig. 8 Composition of aerosol in the Netherlands as a function of size distribution during an episode of eastern wind, February/March 2000.

Table 2 Chemical composition of PM-2.5 in several locations in China as mass concentration/µg m^{-3}. For details, see the article by Zhang et al. elsewhere in this issue. 2000-11 means that the measurement has been carried out in November 2000.

Sites		Year	OC	EC	SO_4^{2-}	NO_3^-	NH_4^+	Mass
Beijing		1994–1995			15.2	5.2	6.5	91.0
Beijing [25]		2000	26.1	5.4	9.9	5.6	4.8	105.0
Pearl River delta [26]		Nov. 2000	19.2	1.7	14.4	2.3	2.4	84.3
Qingdao [14]		1997–1999			16.0	6.9	7.9	49.6
Yangtze delta [24]	Li-an	Nov. 1999	27.7	2.8	15.8	6.7	7.1	73.1
	Changsu	Nov. 1999	40.5	3.0	20.0	11.3	9.4	111.9
	Sheshan	Nov. 1999	33.2	2.0	16.2	9.3	6.6	83.8
Waliguan [29]		1994–1995		0.05–0.6				
Shangdianzi [30]		1999–2000		0.2–0.3				

Note: OC and EC are determined by thermal/optical carbon analysis (TOCA). Ions are determined by ion chromatography (IC).

The total direct effect, backscatter of sunlight, including nitrates and organic compounds is estimated to be about 2 to 3 W m^{-2} for Western Europe. PM-2.5 levels in this area are about 20 to 30 µg m^{-3} and PM-2.5 gives a good indication of the total mass of aerosol in the relevant size fraction for reflecting sunlight.

Soot (black or elemental carbon) plays a totally different role. Soot absorbs incoming solar radiation and heats the atmosphere in this way. The IPCC indicates that this process contributes 0.1 to 0.2 W m^{-2}. East Asia and India are large emitters of soot, due to the use of rather inefficient combustion processes and biomass burning.

The EC values of Chinese cities are probably a factor 2 higher than, e.g., the concentrations measured in Dutch cities (where generally around 1 to 2 µg m^{-3} is measured); see Tables 2 and [4]. The background concentration is of the same order, 0.6 µg m^{-3} or less in Western Europe and China.

The methodologies used to measure black or elemental carbon produce quite uncertain results. Recent intercomparison measurements in Europe indicated that deviations of 100 % or even more occur, even if the same methodology is used. Thus, conclusions cannot be drawn given such large differences until better measurement methodologies are available. The conclusion is warranted that some extra heating due to soot emissions should be taken into account, but this effect would be probably about 0.5 to 2 W m^{-2}.

Aerosols also influence the radiative balance in an indirect way. Cloud formation depends on aerosols. If no aerosols are present, large supersaturation (relative humidity over 100 %) can be observed without droplet formation. However, small particles in different concentrations are present everywhere in the atmosphere. Cloud droplets condense on the aerosols. If few particles (less than 200 cm^{-3}) are available as cloud condensation nuclei (aerosols which act as condensation particles, CCN), large droplets are formed. If a large number of aerosols is present, smaller droplets will be observed. Clouds consisting of many small droplets reflect sunlight much better and hence lead to cooling. The effect is quite large but uncertain. This indirect effect is important if the number of aerosols increases over a low (100 to 1000 particles cm^{-3}) background level. Estimates in Europe have indicated that this effect could be about 0 to –6 W m^{-2} [14]; the same is true for China.

The conclusion must be that, with a large uncertainty due to uncertainties in the measurement methodologies and the lack of relevant data, aerosols cause a net cooling over China (and over many developing countries in the same position) of about –4 to –15 W m^{-2} and are at least as important, but probably much more important, than the effect of the greenhouse gases.

5. CONSEQUENCES OF ABATEMENT OF LOCAL POLLUTION DUE TO AEROSOLS

The lifetime of aerosols is about one week, contrary to most greenhouse gases, which have lifetimes of 10 to several hundreds of years. If the emissions of greenhouse gases would be stopped right now, the effect of the former emissions would be detectable for a very long period. If abatement measures are taken to reduce the quite high aerosol concentrations in fast developing countries, the effects are in place within a week.

The quite serious effects of fine particles on human health, according to the latest scientific findings, will necessitate corrective measures, but one must be aware that these measures will have consequences for other environmental problems:

- Reduction of secondary formed sulfates, nitrates, and ammonium salts will reduce the amount of acid deposition and the impact of eutrophication due to deposition of nitrogen compounds, a beneficial effect.
- The impact on the radiative balance can potentially be enormous. Reduction of the aerosol concentrations to levels in accordance with existing air quality standards (Europe, PM-10, 40 µg m^{-3}) or planned regulations (USA, PM-2.5, 15 µg m^{-3}) would, over a few decades, have potentially at least the same impact as the build-up of greenhouse gases since the industrial revolution.

This observation should not be taken as an admonition not to take measures against high concentrations of fine particles. Rather, it is important that the possible implications of these measures are known.

A good example is acid deposition in former East Germany. Before the reunification with Western Germany, the emissions of precursors of acid deposition were very high, as were also the emissions of basic particles containing calcium carbonate. This calcium carbonate neutralized the acid deposition. The first measures to abate air pollution after reunification were to reduce the emissions of particles. The result was that this region was exposed to acid deposition for a number of years, until the emissions of the precursors were also reduced. It would be important to characterize more accurately the impact of aerosols on the regional climate of developing countries, so the effects of corrective measures can be assessed.

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