

Fine particles in the global troposphere

A review

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ABSTRACT

The available body of data on fine particles ($< 1 \mu\text{m}$ radius) in the troposphere is reviewed in relation to our understanding of sources, sinks and transformation processes of atmospheric aerosols. This review yields the following results. There are no data available to characterize the free troposphere above the boundary layer. Most of the boundary layer data suffer from the lack of a well-defined upper size limit of the samplers at about $1 \mu\text{m}$ radius and concurrent results on total fine particle mass. The fine particle composition data are consistent with our understanding of natural and anthropogenic sources of trace substances, which lead to fine particles. The chemical composition is presented for the three aerosol types: urban, non-urban continental and remote regions. Special attention needs to be focused on fine particle interaction with clouds for three reasons. Fine particles control number- and surface-distribution of the condensed matter; the state of precondensation haze and clouds provide the most important processes of fine particle elimination from the atmosphere. Furthermore, climatic effects of fine particles as expressed by their interaction with atmospheric radiation is strongest during their passage through clouds. Very little is known about the processing of aerosols through clouds. Consequently, there are large uncertainties about their climatic effect and their lifetime in the atmosphere. Finally, no single type of secular trend in particle concentration is found on a global scale. Depending on which component and which geographical region is monitored, both decreasing and increasing concentrations have been measured. The differences are consistent with our understanding of aerosol properties and their gas phase precursors.

1. Introduction

About 80% of the volume of the troposphere lies above the planetary boundary layer ($> \text{ca } 2 \text{ km}$). In this region, important transport and transformation processes are active, determining on a global scale the amount, the composition and the radiative effects of particulate matter. To a reader not familiar with the technical difficulties and analytic limitations, the scarcity of data on concentration, size distribution and chemical composition of particulate matter above the boundary layer might appear astounding. However, a few airborne aerosol studies have been conducted which were directed towards bulk

aerosol composition above the boundary layer (Huebert and Lazrus, 1980; Gaziev et al., 1983, Sebacher et al., 1985). While facing the main problem of collecting enough material for analysis, compromises had to be made concerning the characterization and thus compatibility of samples. If, on the other hand, we include the boundary layer in the scope of the present review, we face a large body of physical and chemical information. Much of these data suffer from serious shortcomings, which prevent or limit their intercomparison in a global synopsis. These shortcomings also constrain or prevent the application of data to radiative transfer and cloud models.

The first part of this paper is dedicated to pointing out some basic guidelines for the study

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of particulate matter relevant to the global tropospheric boundary layer. Arguments will be presented for focusing our attention on submicrometer particles. After a summary of the important source processes for submicrometer particles, their chemical composition is presented for the three aerosol types: urban, non-urban continental and remote marine regions.

A summary of sinks of submicrometer particles leads to a discussion of interaction of small particles and clouds. Finally, our knowledge is reviewed concerning trends in particle concentrations.

2. Fine particles in the troposphere

For studies concerning tropospheric aerosols on a global scale, it is proposed to consider the aerosol as a multiphase system consisting of a carrier gas mixture (air + natural and anthropogenic trace gases) plus solid and liquid particles. When the relative humidity exceeds 100% this system includes cloud elements as another component since the aerosol passage through clouds is a very frequent tropospheric process with serious consequences for the atmospheric lifetime and climatic effects of particulate matter.

The multimodal mass distribution of atmospheric aerosol particles has been demonstrated extensively by Whitby (1978) over a wide range of particle radii and concentration. Fig. 1 summarizes his findings for continental aerosols. Grand average number-, surface- and volume distributions are presented based on the data in Whitby (1978). As the examples in Fig. 2 show, even in remote marine regions, the multimodal character of the aerosol is preserved. At each particle size, one can expect: *external mixtures* in which each particle is one of many pure compounds; *internal mixtures* in which each particle contains a number of mixed chemical compounds or transitional stages in between the extreme types of mixtures. This concept was postulated by Junge (1963) and expanded by Winkler (1973). Hence, for a complete understanding of the atmospheric aerosol, chemically resolved particle size distributions and the state of mixture need to be determined on a global scale.

If we take a second, less sharp look at the multimodal atmospheric aerosol, we distinguish

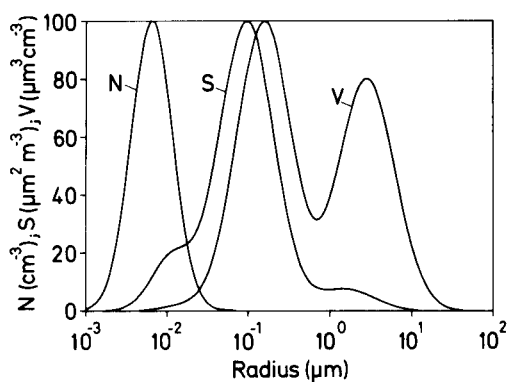


Fig. 1. Grand average number size distribution ($N = dN(r)/d \log(r)$), surface size distribution ($S = dS(r)/d \log(r)$) and volume size distribution ($V = dV(r)/d \log(r)$) for continental aerosols after Whitby's (1978) log normal approximations. 100 divisions correspond to 1.0610^5 cm^{-3} , $638 \text{ } \mu\text{m}^2 \text{ m}^{-3}$, $38.4 \text{ } \mu\text{m}^3 \text{ m}^{-3}$ respectively.

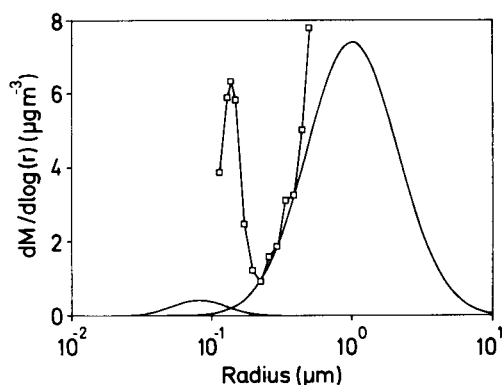


Fig. 2. Bimodal mass size distributions in remote marine regions. (—) log normal distributions fitted to Bigg's (1980) results at Cape Grim. The fine particle mode contains 220 (sulfate)-particles cm^{-3} while the sea salt component has 17 (NaCl)-particles cm^{-3} . (—□—) Clarke's et al. (1987) results for Central Pacific aerosols.

two broad size classes. *Fine particles* (less than $1 \text{ } \mu\text{m}$ radius) differ strongly from *coarse particles* (larger than $1 \text{ } \mu\text{m}$ radius) in terms of their sources and consequently their chemical composition. Atmospheric residence times of particles are strongly dependent on their size (besides other particle properties), as Jaenicke (1978) has pointed out. Fine particles have the longest residence times (on the order of days to a week).

Hence, on a global scale, our attention should be focused on that range.

There are very simple means of eliminating coarse particles from practically any sample flow with cyclones or impactors. However, most of the data on particle composition have been derived from bulk samples without an upper size cut or with size cuts including much of the coarse particle range. On a global scale, marine aerosols are of great importance. In these aerosols, a non-sea salt sulfate component has been recognized (Junge, 1963) which recently has received increasing attention. Evidence indicating the climatic importance of non-sea salt sulfate particles on a global scale is increasing (Charlson et al., 1987). Size distribution data, e.g., Gravenhorst (1978) or Bigg (1980) identify the non-sea salt component at submicrometer radii along with the strong sea salt component. A log-normal approximation of Bigg's results is plotted in Fig. 2. Obviously, the composition of total particle samples will be dominated by sea salt, thus masking effectively the non-sea salt sulfate. The results of Clarke et al. (1987) corroborate an approach of using a radius cut between 0.1 and 0.2 μm when studying non-sea salt marine particles. The fact that he finds much higher non-sea salt masses may depend in part on the different methodologies. In Bigg's studies, the sample preparation for the electron microscope may have eliminated volatile components. Further, the Clarke et al. results were derived in upwelling regions with higher biological productivity.

Ideally, it should be possible to sum the results of specific chemical analyses to equal the total suspended particulate mass (TSP). In the practical case of incomplete analyses, TSP or related aerosol parameters should be available in order to relate the chemical concentrations to the total particulate mass. Only then can aerosol chemistry be properly linked to atmospheric processes, such as radiative transfer or cloud microphysics, which are dependent on all the species that are present. Multicomponent chemical analyses contain much information for source-receptor modeling. Also here, TSP data are essential for a normalization of the chemical results before the multivariate statistical analyses directed towards the apportionment of particle sources. This procedure provides the easiest way of reducing the meteorological variance in a multivariate data set

(Hansson et al., 1984). With today's technology, particulate samples down to a few micrograms can be weighed with electrobalances, piezoelectric balances or β -gauges thus allowing TSP-measurements with about the same time resolution as for chemical analyses (Heintzenberg and Covert, 1987).

3. Sources and growth processes

Directly or indirectly, fine particles are formed by condensation. Here, we disregard the tail of the coarse particle size distributions reaching into the submicrometer range. There are both natural and anthropogenic processes leading to the production of condensable vapors, the most prominent of which being water vapor. Together with other trace gases like H_2SO_4 and ions, homogeneous nucleation can occur below 100% relative humidity, resulting in the formation of new particles. Combustion processes produce large amounts of condensable material, which form primary fine particles in the source (such as elemental carbon) on which vapors from the same source condense (such as high molecular weight aromatic hydrocarbons).

Submicrometer particles grow by means of physical processes that are peculiar to this size range. Coagulation by Brownian motion leads to a transfer of mass from smaller to larger particle sizes while reducing the total number of particles. Condensation to the surface of pre-existing particles leads to particle growth at sizes near the maximum of the surface size distribution. Depending on the hygroscopic particle properties and the relative humidity, varying amounts of liquid water will accrete on the particles, allowing liquid phase reactions which may cause a volume dependent growth. Depending on which process limits the growth rate and on the shape of the pre-existing particle size distribution, different submicrometer-size distributions will be produced (Hering and Friedlander, 1982). In principle, all these processes cause a transformation from external to internal mixtures. Hence, the ultimate stage of an aged, submicrometer particle population should be an internal mixture of all condensable material, reaction products and pre-existing particulate matter in a size region where the mass-transferring processes among different

sizes and removing processes have a minimum. This radius range is between 0.05 and 0.5 μm and has received the name *accumulation mode*.

During cloud events, the source processes of fine particle material can change drastically. Haze particles consisting of submicrometer solution droplets will be diluted by factors of 10^4 – 10^6 after the activation to cloud droplets, with subsequent changes in chemical equilibria. The large surface area of the droplet distribution facilitates the incorporation of soluble gases and the diffusion scavenging of unactivated fine particles. Provided there are sufficient amounts of reactants (such as dissolved SO_2) and oxidants (such as H_2O_2) available and given adequate length of time, stable products will form, which add material to the fine particle population after the dissolution of the cloud. At present, there is considerable uncertainty as to how much of the major aerosol component SO_4^{2-} is produced by one cloud event. In terms of SO_2 -oxidation, figures between 0%/hour, and 1500%/hour, have been reported from different cloud experiments (Daum et al., 1984; Kallend et al., 1982).

4. Fine particle chemical composition

It is very difficult to extract globally representative data on atmospheric particle composition from the literature. The above-mentioned minimum screening of results with a required coarse particle cut-off greatly reduces the data set. It shrinks even further when requiring concurrent data on total fine particle mass (TFP). Following these criteria, only surface measurements were found in the literature. In some cases, TFP-values from related non-chemical data sets had to be used. These data were divided into three aerosol types: urban, non-urban continental and remote marine regions. Only those chemical components were included which were available for all data sets. The results should be used with some reservations. The results on organic carbon come from US regions with photochemical smog problems. NO_3^- and NH_4^+ are volatile components, where the related gas-phase species often were not determined. Hence, the results are not meant to give more than an order of magnitude estimate of natural concentrations or compositions. To some extent, the volatility problem concerns the

halogens as well. However, here, volatile artifact formation on the sample surfaces can lead to chlorine- or bromine-losses at low sample pH (Klockow et al., 1979).

The urban data set comprise 21 experiments in Chinese, European, Japanese and American cities. Table 1 gives the sampling sites. Fig. 3a gives the grand average fine particle composition down to the 6%_{om} (percent by mass) level. Another 14%_{om} were analysed but below this level, 4% by mass were not analysed. All analysed elements with concentrations below the 10%_{om} level are collected on a logarithmic scale in Fig. 3b. This type of diagram also is chosen in Figs. 4b and 5b in order to create a type of chemical signature of the minor chemical components in particles of the three different aerosol types. With the average urban TFP value of 33 $\mu\text{g m}^{-3}$, absolute concentrations can be recalculated.

Under the label "non-urban", results were collected from experiments on the continents but away from large urban or industrial sources. The data were averaged over 35 sets from 16 experiments. The site information can be found in Table 2. Average TFP in non-urban continental aerosols is only a factor of two lower than the corresponding urban value. This result reflects the long residence times of submicrometer particles and the proximity to industrial regions. Consequently, no drastic changes in fine particle composition occur when moving away from the urbanized areas (see Fig. 4a). 2/3 of the fine particle mass is composed of sulfates and carbon compounds while about 15% is nitrogen compounds. 1/4 to 1/6 of the carbonaceous material is black, elemental carbon (EC). It is this minor component that is responsible for most of the absorption of solar radiation in the condensed phase, including in-cloud elements.

The data sets from remote regions include results from 13 experiments. Except for two north polar studies, all data were taken on remote islands or research vessels. The northern and southern hemispheres are covered roughly evenly. Site information can be found in Table 3. For want of free tropospheric data, these mostly marine results are presented as a rough estimate of fine particle composition in the least-contaminated parts of the troposphere.

The average TFP-value is lower by another factor of 3 compared to the non-urban continen-

Table 1. Site information concerning the urban aerosol data set

Site	Reference
New Jersey, USA	Liroy et al. (1985)
Southern California, USA	Appel et al. (1976)
Portage, WI., USA	Spengler and Thurston (1983)
Topeka, KA., USA	Spengler and Thurston (1983)
Harriman, TN., USA	Spengler and Thurston (1983)
Kingston, TN., USA	Spengler and Thurston (1983)
Waterdown, MA., USA	Spengler and Thurston (1983)
St. Louis, MO., USA	Spengler and Thurston (1983), Dzubay and Stevens (1975)
Steubenville, OH., USA	Spengler and Thurston (1983)
Leeds, UK	Clarke et al. (1984), Willison et al. (1985)
Chicago, IL., USA	Scheff et al. (1984)
Berlin, FRG	Lieback and Rden (1983)
Peking, China	Winchester and Bi (1984)
Hamburg, FRG	Heintzenberg and Winkler (1984)
Tokyo, Japan	Yoshizumi and Hoshi (1985)
Stockholm, Sweden	Heintzenberg and Covert (1984)
Detroit, MI., USA	Wolff et al. (1985)
Denver, CO., USA	Countess et al. (1981)
Sao Paulo, Brazil	Orsini et al. (1986)

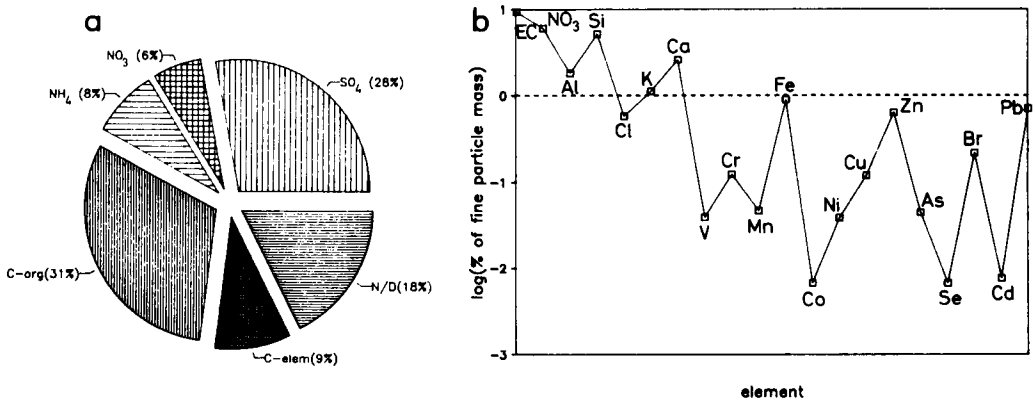


Fig. 3. Average urban fine particle composition (a) down to the 6%_{om} (by mass) level. Average fine particle mass is $32 \mu\text{g m}^{-3}$. $\text{N/D} = 4\%$ _{om} undetermined + 14%_{om} below the 6%_{om} level. (b) Average urban fine particle composition below the 10%_{om} level. The 1%_{om}-line is marked with a dashed line.

tal level. However, only a minority of remote region reports included TFP. Moreover, an average marine TFP-value calculated from surface winds according to the formula given in WMO (1983) might be more representative than the few available observations. On the average, 30%_{om} of TFP remained undetermined, which may be related in part to the uncertainty in TFP itself, or the lack of measurements of silicates,

carbonates and condensed or chemically bound water.

The results shown in Fig. 5a reflect the fact that the fine-particle tails of coarse-mode sea salt size distribution and to a lesser extent crustal particle component (cf. Fig. 1) affect the fine particle composition in remote regions. Nevertheless, even here sulfate compounds are a major component. About 1/5 of the fine particle mass is

Table 2. Site information concerning the non-urban continental data set

Site	Reference
Northeastern, USA	Ferek et al. (1983)
Velen, Sweden	Lannefors et al. (1983a)
Juréia, Brazil	Orsini et al. (1986)
Lancaster, UK	Harrison and Pio (1983)
Pennsylvania, USA	Lewin et al. (1986)
Haverah Park, UK	Willison et al. (1985)
Arizona, USA	Macias et al. (1981)
Tennessee, USA	Stevens et al. (1980)
Virginia, USA	Tuncel et al. (1985), Ferman et al. (1981)
Minnesota, USA	Pratt and Krupa (1985)
Wisconsin, USA	Pratt and Krupa (1985)
Kentucky, USA	Shaw and Paur (1983)
Delaware, USA	Wolff et al. (1986)
"Non-urban", USA	Altshuller (1976)

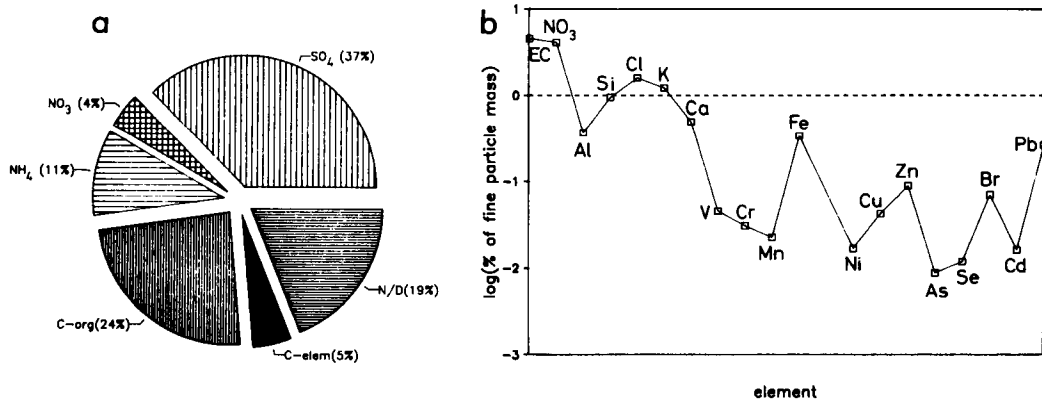


Fig. 4. (a) As Fig. 3(a) but for non-urban continental fine particles. Average fine particle mass is $15 \mu\text{g m}^{-3}$. $\text{N/D} = 14\%_{\text{om}}$ undetermined + $4\%_{\text{om}}$ below the 6% level. (b) As Fig. 3(b) but for non-urban continental fine particles.

Table 3. Site information concerning the remote regions data set

Site	Reference
Hawaii	Duce et al. (1983)
Bermuda	Wolff et al. (1986), Duce et al. (1983)
Samoa	Duce et al. (1983)
Bonin Islands	Youshizumi and Asakuno (1986)
North Atlantic	Winkler (1975)
South Atlantic	Mészáros and Vissby (1974)
Greenland	Flyger and Heidam (1978)
Pacific Ocean	Saltzman et al. (1986)
Tasmania	Andreae and Barnard (1981), Ayers et al. (1986)
Namibia	Annegarn et al. (1983)
Arctic Ocean	Lannefors et al. (1983b)

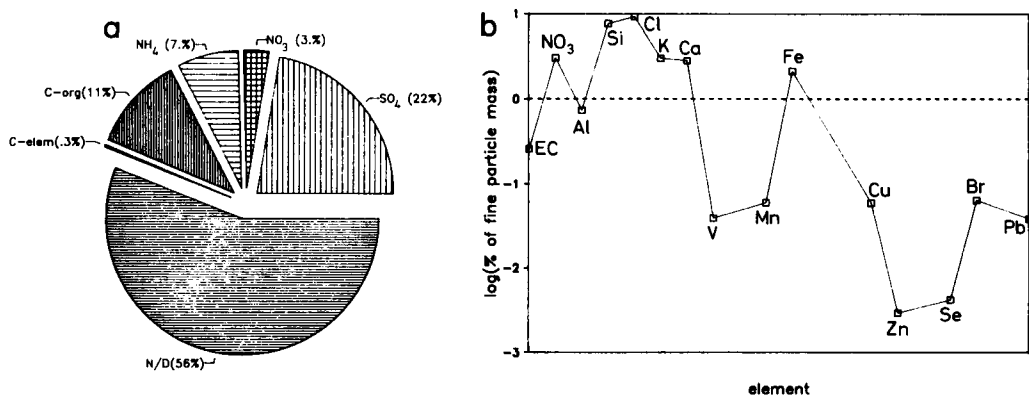


Fig. 5. (a) As Fig. 3(a) but for remote regions fine particles. Average fine particle mass (TFP) is $4.8 \mu\text{g m}^{-3}$. N/D 30% undetermined + 26%_{om} below the 6%_{om} level. (b) As Fig. 3(b) but for remote regions fine particles.

sulfur compounds. Organic carbon is the next most important component with about 11% of TFP. EC as the most clearly identifiable anthropogenic component exhibits only 0.3% corresponding to an average concentration of 10 ng m^{-3} .

After eliminating data from the polar regions and marine areas up to 1000 km downwind of a continent from our data set, average fine particle composition in the remote marine boundary layer can be calculated. It has not been shown that the average sea water composition can be applied to all components in submicrometer sea salt particles. Of the two major sea salt components Na⁺ and Cl⁻ used as sea water references, the former is often not included in multielement studies while the latter often yields erroneous results because of chemical artifacts. However, chemically, we can be confident that Na⁺ and SO₄²⁻ from sea water are conserved in the marine aerosol. Hence, additional non-sea salt SO₄²⁻ was derived for those experiments including Na. In the fine particle range we derive a global average concentration of $0.5 \mu\text{g/m}^3$ of non-sea salt sulfate.

If we look at the concentrations of the minor components in the 3 aerosol types (Figs. 3b–5b), we find a general downward trend with increasing atomic mass. In the 2 continental aerosol types, however, the metals iron, zinc and lead strongly interrupt this trend with concentrations in the range 0.1–1%. This chemical signature of minor components in fine particles becomes quite different when analyzing remote marine aerosols.

The relative contribution from anthropogenic components like EC and Pb is strongly reduced, while crustal and marine components like Si, Cl, Ca and Fe become dominating. They even contribute strongly to TFP as seen in the unspecified but analyzed part (26 units of the 56% N/D) in Fig. 5a.

5. Sinks for fine particles

Any process which causes a particle to cease to exist as a separate entity, we define as a sink. Of the two most important of the dry sink mechanisms, coagulation and sedimentation, the latter can be neglected in the fine particle range because of their fall speeds being less than 0.01 cm s^{-1} . Coagulation through thermal diffusion, on the other hand, becomes increasingly effective with decreasing particle size. However, only diffusion to the earth's surface eliminates particles from the atmosphere, otherwise no aerosol mass is lost. Coagulation is often split up into the interaction of particles with similar sizes (self-coagulation) and coagulation with particles of different sizes. In order to increase the particle size up to the stable $0.1 \mu\text{m}$ radius region (confirm section three), a large number of self-coagulation steps would be required for the small primary nuclei of less than $0.01 \mu\text{m}$ radius from natural and anthropogenic sources. Coagulation with pre-existing larger accumulation mode particles is much more effective. As a result, the

particle concentration in this size range determines the turn-over time with respect to coagulation of the primary nuclei (Whitby, 1978). This turn-over time is defined as the ratio of the total mass of nuclei divided by their mass flux to a pre-existing population due to coagulation. Coagulation with coarse particles, on the other hand, is negligible (Whitby, 1978). At low pollution levels, coagulation yields turn-over times between 1 hour and 100 days for fine particle sizes between $0.001\ \mu\text{m}$ and $0.1\ \mu\text{m}$, respectively (Jaenicke, 1978).

In order to arrive at realistic average age of accumulation mode particles in the atmosphere (3 to 30 days), cloud and precipitation processes have to be taken into account (Jaenicke, 1978; Ogren and Charlson, 1983). Even here, the hygroscopic particle properties are expected to affect their activation as cloud droplets (Köhler, 1936; Jensen and Charlson, 1984). The incorporation of fine particles by either nucleation in or diffusion to cloud droplets can be extremely effective. 12-hour median concentrations of $0.1\ \text{particle cm}^{-3}$ have been measured in Arctic fogs as compared to out-of-cloud concentrations of several hundred cm^{-3} . Depending on the hygroscopic particle properties, the type of clouds and their frequency of occurrence, fine particle lifetimes from shorter than a day in tropical climates to several weeks in the winter Arctic are expected (Ogren and Charlson, 1983). The discussion of fine particle sink processes emphasizes their rôle as an intermediate reservoir in the processing of trace substances through the atmosphere.

6. Fine particles and clouds

We begin our discussion of the interaction between fine particles and clouds in the *precondensation stage*, which we define as the humidity range between the deliquescence points of the major hygroscopic aerosol components (60–80%) and 100% relative humidity. Hygroscopic particle behavior has been investigated for many years. However, there are few data available, which are based on in situ experiments (as compared to measurements on particle deposits). The humidograph technique developed by Covert et al. (1972) allows bulk in situ measurement of hygroscopic particle properties without yielding

results on the state of mixture (confirm Section 2). This state is lost when depositing particle samples prior to analysis except when studying deposited single particles with an electron microscope.

Table 4 summarizes the available in situ results on the state of mixture and hygroscopic particle properties (Covert and Heintzenberg, 1984; Heintzenberg and Covert, 1987). There is a general agreement that the so-called Arctic haze phenomenon is caused by continental aerosols coming from anthropogenic midlatitude sources, Stonehouse (1986). Consequently, the only available non-urban continental results are based on two samples taken in Arctic haze on Spitsbergen. The mass increase by a factor of two derived for a particle humidification to 90% is consistent with results from measurements on deposits and other in situ measurements (Covert et al., 1972; Winkler et al., 1981). The lack of growth of elemental carbon strongly suggests an external mixture of hydrophobic soot and hygroscopic particles in continental aerosols. Since elemental carbon comprises only some 5% of TFP the in situ results are consistent with results from electron microscopic studies indicating more than 80% of the fine particles being hygroscopic (Okada, 1983).

On the other hand, soot has been measured in rain, snow (Clarke and Noone, 1985) and cloud water (Ogren and Charlson, 1984). At present, it is unclear by which process hydrophobic material such as soot is processed through clouds. This question is of importance over industrialized regions as is illustrated by the results of model calculations of heating rates in a stratus cloud in Fig. 6. Heating rates in clouds vary over a factor of ~ 2 depending on how the absorbing particles are distributed in the cloudy air. Least absorption

Table 4. *Fractional increase of fine particle mass, sulfur, soot (EC) and trace metals in the radius range, $0.26\ \mu\text{m} < R < 1.0\ \mu\text{m}$ due to hygroscopic growth when humidified to 90%*

aerosol type	mass	S	Fe	Pb	EC
urban	2.1	2.4	N/D	3.9	1.1
non-urban continental	2.0	2.8	2.1	2.7	0.94

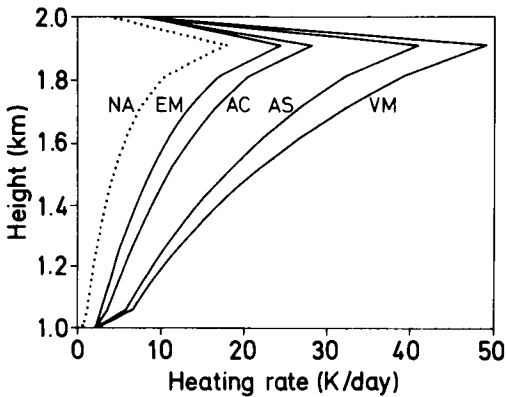


Fig. 6. Heating rates due to the absorption of solar radiation calculated for a stratus deck with an optical density $\tau_{0.1} = 25$ between 1 and 2 km altitude over a surface with albedo $A_s = 0.2$ for a standard mid-latitude winter atmosphere (MLW) and a sun position given by $\mu = 0.7$. The volume mixing ratio of light absorbing material and cloud water is $V = 10^{-4}$. NA = reference case with no soot, EM = external mixture of soot and droplets, AC = internal mixture of absorbing core and a water shell, AS = internal mixture of an absorbing shell and a water core, VM = volume mixture with "dissolved" soot throughout each droplet.

occurs in hydrophobic external mixtures. The EC volume mixing ratio $V = 10^{-4}$ in the model calculation corresponds to heavily polluted clouds such as expected over Central Europe in winter.

Most clouds evaporate without undergoing precipitation. They leave behind mainly fine particles and a few coarse ones, which to some extent were incorporated in the droplets and consequently affected by processes in the droplets. We now have an instrument available which simulates an evaporating cloud allowing the study of particle processing through this complicated multiphase system. This counterflow virtual impactor cloud probe (CVI) isolates the non-volatile trace substances in the droplets in the airborne state and can be connected to a wide range of particle analyzing systems (Ogren et al., 1985). With the CVI, it is now possible to see what fine particle population is released by an evaporating cloud. First results on particle-size distributions after cloud detrainment were described in an stratocumulus experiment at a rural mountain site in central Sweden. Site and experiment are described in Johansson (1987).

Size distribution measurements after the CVI show mass size distributions, which are similar to non-urban continental data given by Whitby (1978) suggesting a fine particle population, which leaves a single non-precipitating cloud rather unaffected.

7. Trends in fine particle concentration

The amount of total suspended material or related parameters has been monitored for several decades at many European and US sites. Because of ill-defined upper size-cuts in the samplers (cf. Section 2), the results are often hard to evaluate and even harder to compare. However, there seems to be a general trend in TSP measured in urban areas. Since the middle of the fifties, TSP-values have gone down in many locations by a factor of two to three. The increase in paved roads and a reduction in the emission of flyash have been given as explanations for the reduction of emissions to rural areas. Since the particle sizes of flyash are in the short-lived coarse particle range, these concentration decreases have little significance on a global scale where trends in TFP or related parameters are most interesting. Jaenicke has evaluated several TFP-related time series (PROMET, 1986). Kaminski and Winkler (1986) have presented a detailed trend analysis of their fine particle measurements. In both reports, statistically significant upward trends of fine particle related parameters are found in Germany over the last ten years. On a global scale, time series related to fine particles do not exist. The reason for the observed increase in fine particle mass are suspected to lie in the source mechanisms of this type of aerosol material. Upward trends in gas phase precursors especially sulfur- and nitrogen oxides will cause an increase in the amount of condensed material in the particulate phase (Möller, 1984).

The material of this secondary aerosol is practically transparent in the solar spectral range. In order to estimate possible climate effects of such trends, we need to know concurrent trends in light absorbing fine particle material such as elemental carbon. A time series of this parameter in marine air has been started about 5 years ago at the Australian baseline station at Cape Grim

(Heintzenberg, 1985). Up until 1987, no significant trend in light absorbing matter was found in the time series. No such data are being acquired currently in the northern hemisphere.

8. Conclusions

The available body of data on fine particles in the troposphere has been reviewed in relation to our understanding of sources, sinks and transformations processes of atmospheric aerosols. A number of conclusions can be drawn from this review. There are no data available to fully characterize the free troposphere above the boundary layer. Most of the boundary layer data suffer from the lack of a well defined upper size limit of the samplers and concurrent results on total fine particle mass. Fine particle composition data, which pass such a basic screening are consistent with our understanding of natural and anthropogenic sources of trace substances, which lead to fine particles.

Special attention needs to be focused on fine particle interaction with clouds for two reasons. Precipitating clouds provide the most important processes of fine particle elimination from the atmosphere. Furthermore, climatic effects of fine

particles as expressed by their interaction with atmospheric radiation is strongest during their passage through clouds (Twomey, 1974). Cloud occurrence and cloud albedo are determined by the number, size, hygroscopic and light absorbing properties of fine particles. Very little is known about the processing of aerosols through clouds. Consequently, there are large uncertainties about their climatic effect and their lifetime in the atmosphere. Finally, no single type of secular trend in particle concentration is found on a global scale. Depending on which component and which geographical region is monitored, both decreasing and increasing concentrations have been measured. The differences are consistent with our understanding of aerosol properties and their gas phase precursors.

9. Acknowledgements

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