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Chemically resolved submicrometric size distribution and external mixing of the Arctic haze aerosols

By JOST HEINTZENBERG, *Department of Meteorology*, University of Stockholm, Arrhenius Laboratory, S-10691 Stockholm, Sweden* and DAVID S. COVERT, *Department of Environmental Health, University of Washington, Seattle, WA 98195, USA*

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ABSTRACT

Atmospheric aerosol particles were collected with filter and impactor samplers at Ny-Ålesund, Spitzbergen during an Arctic haze event in the Spring of 1983. Additional integral aerosol parameters particle light scattering coefficient and total number concentration were also measured. The samples were analyzed for mass, elemental and soot content. With the exception of chlorine that was due to local marine sources, concentrations of the chemical species were all highly correlated with each other and with total mass concentration, light scattering and total number concentration. Mass concentration varied between 4 and 12 μg m^{-3}; other species showed values within the range reported previously under similar conditions. The impactor and total number concentration data were inverted to yield size distributions for the size range 0.01 to 1.0 μm radius. The mode of the total and elemental mass size distributions was in the range 0.13 to 0.26 μm with the exception of soot, which had a mode at slightly less than 0.13 μm. The main mass of the particulate material had hygroscopic properties similar to that previously observed for urban or continental aerosols. However, soot was observed to be associated with particles that were hygrophobic and it was concluded that it was externally mixed. Overall, the aerosol had properties indicative of one that had remained relatively unchanged after transport from its source region.

1. Introduction

In recent years, our knowledge about the phenomenon of Arctic haze has increased substantially, partly due to long term monitoring and regular point experiments, partly because of intensive multinational measuring campaigns covering large areas. A review of the availability body of data from the European Arctic can be found in the work of Heintzenberg et al. (1986).

Two of the major tasks remaining are the identification of distant sources and mechanisms of aerosol formation and removal in the Arctic. One way of approaching these problems is through the measurement of the chemically resolved submicrometric size distribution of the haze particles. Shaw (1983) and Heintzenberg et al. (1986) have shown that the shape of the Arctic size distribution contains information about particle formation and aging processes, especially when particles with radii, R, below 0.1 μm are included in the analysis. When pattern recognition techniques were applied to chemically resolved size distributions, Martinsson et al. (1984) found significant differences in elemental size distributions depending on source origin.

Aerosol transformation and removal processes are strongly dependent on the hygroscopic properties of particles. Water uptake in the state of precondensation and in cloud processes determines atmospheric lifetimes of climatically important aerosol components such as elemental carbon (C°), (Ogren and Charlson, 1983). So far, only one data set concerning the hygroscopic behaviour of C° exists (Covert and Heintzenberg, 1984), indicating that in the urban environment

* Contribution no. 561.

soot is contained in particles that are relatively hygrophobic.

In March 1983, one of the authors (DSC) conducted a field experiment at Ny-Ålesund, Svalbard, to collect size segregated aerosol samples for subsequent chemical analyses. A detailed description of the measuring site and the measures to minimize local contamination can be found in Heintzenberg et al. (1983). In the course of two weeks, six sets of samples with four size channels each were collected. In the following we describe the experiment and evaluate the results of the chemical analyses with an inversion algorithm to recover the chemically resolved mass size distribution of the Arctic haze aerosol in the radius range 0.01 to 0.5 μm. Two additional samples were taken at a controlled humidity of 90% to determine hygroscopic properties of the different chemical compounds in the aerosol.

2. Experimental

An air sampling station operated by the Norwegian Institute of Air Research at Ny-Ålesund, Spitzbergen was used as the site for the present experiment. The month of March, 1983 was chosen for the experiment because it usually exhibits the annual peak in Arctic haze burdens (Heintzenberg et al., 1986). The experimental setup is nearly identical to the one used in an urban aerosol sampling experiment (Covert and Heintzenberg, 1984; Heintzenberg and Covert, 1984). Fig. 1 shows the complete aerosol sampling and recording train. We did not use the diffusion battery of the previous study and the last 2 impactor stages were combined to increase sample masses for that size range and to reduce the effect of sizing errors introduced by particle bounce-off. Thus, the information content of the combined data set, was reduced and the inversion results discussed below exhibit a reduced size resolution compared to the urban results of Heintzenberg and Covert (1984). As supporting continuous data, we recorded the particle light scattering coefficient at 0.55 μm wavelength with a nephelometer (σ_{sp}), Aitken nuclei concentration (CNC) with a TSI 3020 counter, wind direction and relative humidity. Sample collection was controlled by the CNC and wind direction sensor to minimize contamination from local sources.

Gravimetric, PIXE (particle induced X-ray emission) and photometric analyses for particulate mass, elemental composition and soot were performed on the same particulate deposits of each sample. The procedures are described in Heintzenberg and Covert (1984). It should be stressed that, in order to reach the necessary gravimetric detection limits of 0.5 μg/sample, electrobalance weighing was done at the sampling site.

3. Primary results

In this section, we report primary results, i.e., direct recordings of atmospheric or aerosol parameters, and the results of the chemical analyses of the total submicrometric aerosol samples. Size segregated aerosol composition in terms of hygroscopic properties and mass size distribution will be discussed subsequently.

Six samples were collected during the period 13 March to 28 March, 1983. In Fig. 2, the results from the mass and chemical analysis of the filter samples of total submicrometric aerosol are presented in the form of a time series. Average values for σ_{sp} and CNC over the filter sampling periods are presented with the filter results. Each parameter has its own scale labelled with minimum and maximum values on the left-hand side of the graph. The time series exhibits a very consistent picture of uniform aerosol composition. Except for chlorine, all aerosol components and integral parameters such as CNC and σ_{sp} are highly correlated, indicating common, distant large sources and common transport paths and processes except for the sea salt derived chlorine.

A trajectory analysis reveals the reasons for the measured temporal development of particle concentrations and composition changes. The first sample was taken during the final stage of the severe haze period analysed by Raatz (1985) with air pollutants being transported rapidly from Eurasian industrial sources across the Arctic. During sampling events 2 and 3, surface and 850 mb back trajectories provided by Raatz pass over the Greenland sea, the northern part of the Norwegian sea and the Barents sea before reaching the sampling site thus explaining the low mass concentrations and the peak in chlorine

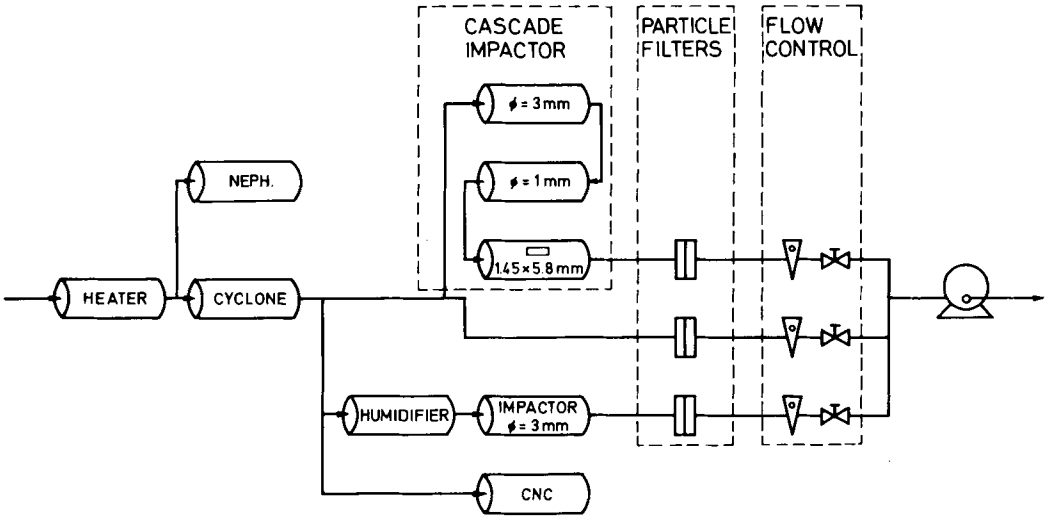

Fig. 1. Aerosol sampling and recording train.

concentrations. Sulfate concentrations measured by Iversen and Joranger (1985) between 1–20 March at the same site confirm our results. Samples 4–6 again were characterized by direct flow from the direction of Eurasia with trajectories originating from the Taymyr Peninsula.

4. Hygroscopic aerosol properties

The hygroscopic behavior of aerosols is described by the concept of a mixed aerosol introduced by Junge (1963) and expanded by Winkler (1973). The two extreme cases are internal mixtures in which each particle contains a number of homogeneously mixed chemical compounds, and external mixtures in which each particle is a pure compound yet the particle population consists of a number of compounds. Any intermediate degree of mixing is possible and we expect any aerosol generation and ageing process in the real world to produce internal mixtures to some degree.

Covert and Heintzenberg (1984) developed a method of collecting size segregated samples of particles with parallel impactors at two controlled humidities as means of determining the degree of mixing of soot and hygroscopic compounds in atmospheric aerosols. Their results were based on the measured change in relative amounts of the total mass and several elemental components of particulate matter in size intervals above and below a radius of 0.26 μm that was caused by an RH change from about 30% to 90%. The urban aerosol samples collected in Stockholm in 1982 showed that soot was associated with relatively hygrophobic particles rather than the hygroscopic particles that comprised the majority of the particulate mass. Thus, the aerosol was largely externally mixed with respect to those two types of particulate matter. In the experiment in Ny-Ålesund in 1983, that same sampling protocol was repeated under Arctic haze conditions to determine the effect of ageing on internal mixing. Six sets of samples were collected during the two week period but only two complete data sets were obtained. In samples 1 through 3, one or more of the size segregated soot samples were near or below the detection limit of the soot photometer. Additionally, sample 1 was considered contaminated because of high soot masses in the Aitken size range (<0.1 μm radius) and inconsistent results of the metal analysis. Sample 4 was flooded because of a malfunctioning humidity control.

The results are presented in two ways in Tables 1a, b. First, the measured concentration of total or elemental particulate mass in the size range $0.26\ \mu m < R < 1.0\ \mu m$ after humidification is expressed as a fraction of the respective mass in that size range at low RH. Mathematically and conceptually this is the more direct result but it is

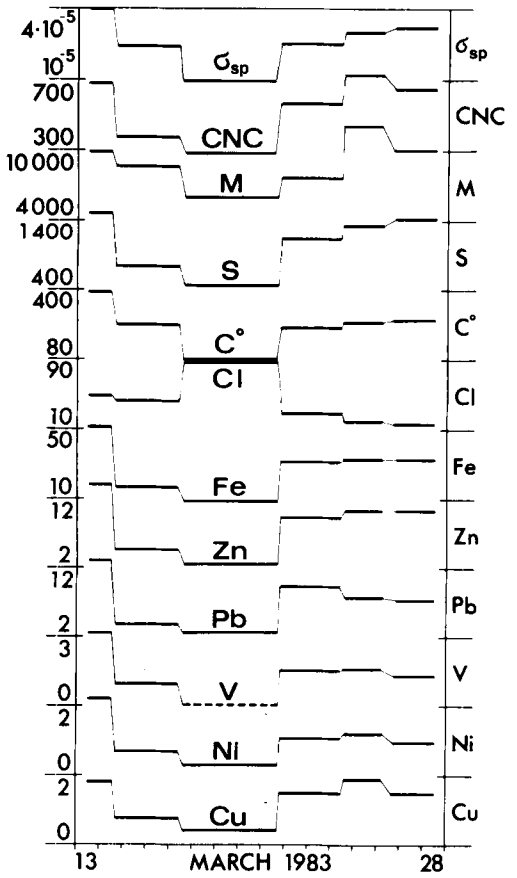

Fig. 2. Time series of physical and chemical aerosol parameters during the Arctic aerosol experiment 13 March to 28 March, 1983. σ_{sp} = light scattering coefficient at 0.55 μm in m^{-1}. CNC = total aerosol number concentration in cm^{-3}. C° = elemental carbon. Each time series has its own scale. A dashed line stands for a value of one half the detection limit as mass concentration. Gravimetric and chemical analysis in ng m^{-3}.

dependent on both the amount of hygroscopic growth associated with an aerosol species and the initial or dry size distribution of that species. It is useful, however, for visualizing the relative changes in mass and elemental concentration that will occur in the size range above the submicrometric mode of the size distribution upon humidification as occurs in the atmosphere. In the second representation of the data, the measured increase in concentration of total or elemental

Table 1a. *Fractional increase of atmospheric aerosol particulate mass, sulfur, soot (C°) and trace metals in the radius range, 0.26 μm < R < 1.0 μm due to hygroscopic growth when humidified to 90% RH*

Sample	Mass	S	Fe	Pb	C°
Ny-Ålesund 1983					
#5	1.8	2.1	2.0	2.4	0.91
#6	2.2	3.4	2.1	3.0	0.96
average	2.0	2.8	2.1	2.7	0.94
Stockholm 1982					
average	2.1	2.4		3.9	1.10

Table 1b. *Fraction of atmospheric aerosol particulate mass, sulfur, soot and trace metals removed from the radius range, 0.13 μm < R < 0.26 μm by hygroscopic growth when humidified to 90% RH*

Sample	Mass	S	Fe	Pb	C°
Ny-Ålesund 1983					
#5	0.48	0.50	1.25	0.8	−0.15
#6	0.62	0.81	1.3	1.1	−0.03
average	0.55	0.65	1.2	1.0	−0.06
Stockholm 1982					
average	0.63	0.52		1.06	0.06

particulate mass in the size range 0.26 μm < R < 1.0 μm is expressed as a fraction of the respective mass in the size range 0.13 μm < R < 0.26 μm at low RH. This is the next lower size range that was sampled and encompasses the mode of the submicrometric size distribution for the measured elements with the exception of C°. The basis for this representation is that at 90% RH, the amount of hygroscopic growth that can be expected for any realistic atmospheric aerosol is less than a factor of two in radius (Tang, 1980; Orr et al., 1958). Thus, any additional mass that was observed in the size range, 0.26 μm < R < 1.0 μm, could have come only from the range 0.13 μm < R < 0.26 μm and not from sizes smaller than 0.13 μm. This provides a more bounded result that is dependent primarily on hygroscopic properties of the particles in the source size range and much less on their size distribution since this varies less within these limits than over the larger range 0.13 to 1.0 μm.

It is this result that should be used for comparing hygroscopic properties of different elements or the degree of internal mixing for aerosol samples collected at different times or locations.

At the bottom of the tables, the averaged results obtained in Ny-Ålesund are compared with data from samples collected similarly in urban Stockholm. The results from Stockholm are based on 8, 7, 2 and 2 samples for particle mass, soot, sulfur and lead, respectively. With a precision of 25% we are confident in the data in Tables 1a and 1b and in the comparison with our previous results.

The data show that, with the exception of the results for lead as expressed in term of the mass increase above 0.26 μm, the size and hygroscopic properties of the urban samples and the Arctic samples agree within the experimental precision. That exception for lead can be interpreted as a difference in its size distribution rather than in the hygroscopic properties of the particles with which it was associated. Lead was more broadly distributed at sizes larger than 0.26 μm in the Arctic samples. The amount of growth of the particles in the 0.13 μm to 0.26 μm range with which lead was associated was about the same in both cases. Hence, our Arctic haze results indicate an external mixture consisting of hygrophobic particles in which the soot was contained on one side and hygroscopic particles containing sulfur and metals on the other. Admittedly, more experimental evidence is needed before drawing general conclusions about the hygroscopic properties of Arctic haze, or other aged aerosols. However, these first results are consistent with all other physical and chemical data suggesting that Arctic haze is formed from mid-latitude aerosols which maintain their anthropogenic characteristics even after 5000 km or 5 days of transport from the source regions.

Since sampling was stopped any time the wind direction or CNC indicated the possibility of local contamination from combustion sources and since the levels of soot observed by other studies in the Arctic are of the same magnitude as ours, we believe that the results are valid for these two samples at least. It is remarkable that particles can retain their initial properties in the face of long ageing times but this is consistant with the coagulation lifetime model of Ogren and Charlson (1983) for just these conditions.

5. Inversion results and discussion

The most common way of presenting mass size distributions from multi-stage cascade impactors are plots of mass concentration for each impactor stage, arranged in order of increasing particle size. Pacyna et al. (1984) reported their size differentiated aerosol composition data from Ny-Ålesund during the same period as our experiment in this form. However, this way of presenting size distributions disregards the non-ideal collection efficiencies of impactors and exhibits open lower and upper limits of the distribution. Consequently, comparison of data from different samplers becomes cumbersome and uncertain. As in our urban aerosol experiment, we inverted the primary set of physical and chemical data to recover the underlying mass and chemical size distributions. This inversion procedure is necessary because of the non-ideal collection characteristics of the aerosol instruments. Each of the sampling channels has some cross-sensitivity in the neighboring channels, some of the instruments cover the radius range 0.005 to 1 μm. Details of the inversion algorithm are given in Heintzenberg (1978). For the inversion process, the collection efficiencies of each sampling channel have to be known with high accuracy. Hence the samplers were calibrated with monodisperse aerosols of known composition (Heintzenberg and Covert, 1984).

As a means of replacing the information of the missing diffusion battery in the Aitken size range, the condensation nuclei counter was added to the data set for the inversion. A prerequisite for this step was a determination of the counting efficiency throughout the submicrometric radius range. Agarwal and Sem (1980) have presented at efficiency calibration below 0.05 μm. We extended their calibration up to 2.8 μm radius and found counting efficiencies of $1 \pm 5\%$ above 0.05 μm radius. Hence, the TSI-3020 can be used as a total particle counter in the whole fine particle range. Before joining the CNC with the chemical mass data, the CNC-counting efficiency was converted to a mass collection efficiency by applying a factor of $\frac{3}{4} \cdot \pi \cdot r^3 \cdot \rho_p$ to the counting efficiencies. This transform is equivalent to applying an efficiency of unity to the number counting efficiency and allows the CNC data to be applied directly to the mass data without

transforming to a number distribution. This factor implies the assumption of spherical particles with the density ρ_p. For ρ_p, we assumed the value of 1 g cm^{-3}, the same as that of the calibration aerosols.

The mass collection efficiencies of the five sampling or measuring channels are plotted in Fig. 3. Obviously, the CNC is very sensitive to particulate mass in the Aitken range, and can be used in the inversion to prevent the build-up of mass artifacts at the lower size end where none of the other channels exhibits sufficient sensitivity or size discrimination.

The information content of the CNC data was explored with the gravimetric data set of sample 4. The mass data were inverted without the CNC results and with CNC results assuming varying relative errors in the CNC measurements as illustrated in Fig. 4. Without the CNC information the inversion builds up unrealistic masses below 0.05 μm radius because none of the impactor channels provides sufficient size discrimination below that radius (see Fig. 3). Even allowing for several hundred percent measuring error by the CNC the build-up of mass artifacts in the Aitken range is effectively suppressed.

The problem of artifacts in inversion results arises as well when inverting the chemical mass data. However, strictly speaking, the CNC results can only be used to constrain the total

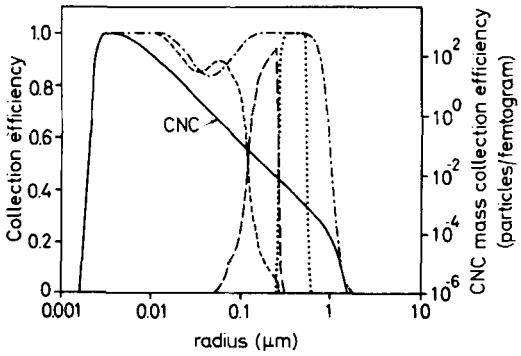

Fig. 3. Size dependent mass collection efficiencies of the 5 channels in the Ny-Ålesund aerosol sampling and recording train. (—·—) total filter. (——) CNC counting efficiencies converted to mass collection efficiencies. (— —) back-up filter plus first impactor stage. (----) second impactor stage. (·····) third impactor stage. Note that a cyclone determines the upper cut-off of all instruments.

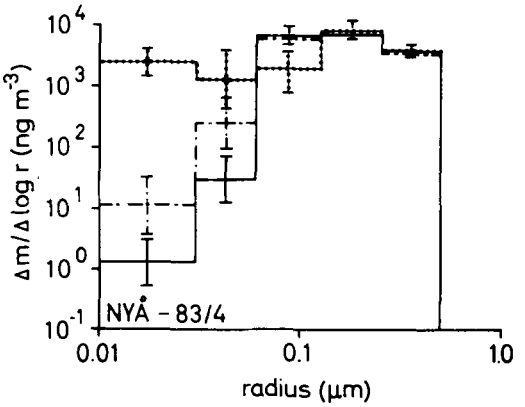

Fig. 4. The influence of the information content of the CNC measurements on the inverted mass size distribution for sample 4. (----) inversion without CNC data, 20% measuring error in the gravimetric data. (—·—) with CNC data having 338% measuring error. (——) with CNC data having 50% measuring error.

particle number as mass. To improve the quality of the inversion of the chemical data, the approach of applying total number concentration was extended to the chemical components by the following assumption. A total particle number was apportioned to the different chemical species according to the mass fraction of the respective species:

$$N_{i,j} = \mathrm{CNC}_i \cdot m_{i,j}/M_i,$$

with M_i being the gravimetric result for the total filter, $m_{i,j}$ the mass concentration of component j on the same filter, CNC_i the average total number concentration for sample i and $N_{i,j}$ the calculated number concentration for component j in sample i. This procedure presumes that the size distributions of the different chemical species have the same shape. Previous urban (Heintzenberg and Covert, 1984) and Arctic haze results (Heintzenberg et al., 1981) show that this is approximately the case. In order to allow for the possibility of some variation the inverted size distribution of the different species, $N_{i,j}$ was allowed to have a measuring error of 338%. This value corresponds to a variation in the mass median radii of the different components by 50% while still suppressing inversion artifacts at the small sizes (see Fig. 4).

The inversion results reported below are based on 20% measuring errors in the analysed masses and 338% in the calculated $N_{i,j}$ (corresponding to

a 50% uncertainty allowed in mass medium radii). The total mass size distributions are based on one common 20% measuring error.

The gravimetric measurements yielded results above the detection limit for all 6 samples. Furthermore, we feel that the chemical contamination in sample one did not influence the total mass which is supported by the nephelometer results of sample one which reflect total mass concentration. Hence, we present total mass size distributions for all samples in Fig. 5. Mass median radii in all cases are in 0.13–0.25 μm range in accordance with previous results (Heintzenberg, 1980). The areas under the histograms reflect the temporal development of the total mass as discussed in connection with Fig. 2. The passage of marine air masses over the station lead to a clearing of the size distribution beginning at the smallest sizes (sample 2), leaving a broad low level mass distribution similar to the results of Jaenicke and Schütz (1982) on Arctic aerosols during the clean summer months (sample 3). During the latter half of the sampling period the concentration increased again particularly at sizes less than 0.13 μm. No significant particle masses were found below 0.03 μm in any measurement. Samples 4–6 cover the second haze period of the experiment. We averaged the results for these samples to derive a representative chemical size distribution for the period 21–28 March. Inversion of the sample averages yielded the chemically resolved mass size distribution shown in Table 2 and Fig. 6. A log/log scale was chosen in Fig. 6 to be able to show the small absolute masses in the Aitken range. Mass median radii for most of the chemical components are close to 0.1 μm. Only C° has a slightly smaller median and Fe exhibits a continuous mass increase throughout the investigated size range. These findings are consistent with our understanding of the different aerosol sources in the fine (<1 μm radius) and coarse (>1 μm radius) particle range. C° is combustion derived with primary particle sizes around 0.01 μm while Fe in the Arctic is mainly crustal material which lies in the coarse particle range. All species were present at measurable mass concentrations in the Aitken range.

In Table 2, the analysed mass concentrations in each of the 5 histogram columns are summed for a comparison with the inverted gravimetric results. Below 0.1 μm radius 30–50% of the weighed mass is accounted for by the chemical analysis. At the same time, the inversion results themselves exhibit combined standard deviations which are much larger than the discrepancy between mass determinations. Hence, we refrain from speculating about missing chemical components in the Aitken range. Near the maximum of the mass distribution (columns 4 and 5) 90% and 75% respectively of the gravimetric mass is accounted for by the chemical analyses. S interpreted as $SO_4^=$ is the predominant species on a mass basis.

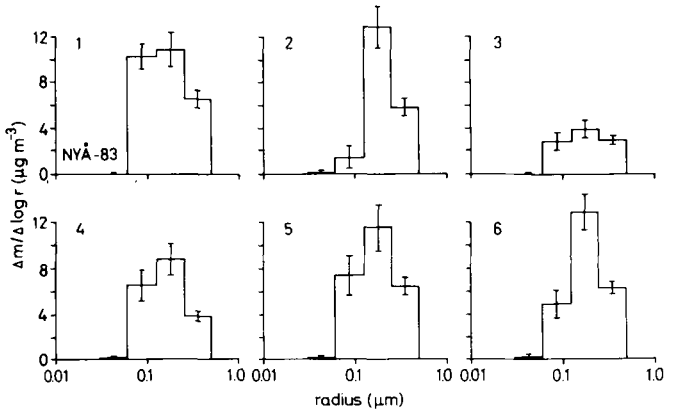

Fig. 5. Mass size distributions of Arctic haze at Ny-Ålesund, Spitsbergen. The 6 samples cover the period 13–28 March 1983. The error bars show the combined effects of measuring plus inversion errors.

Table 2. *Size-segregated submicrometric aerosol composition from inversion of average sample data of samples 4–6 in ng m^{-3} per histogram size class (μm)*

Size class (μm)	0.01–0.03	0.03–0.06	0.06–0.13	0.13–0.25	0.25–0.50
S	1.8E + 0	2.6E + 1	7.2E + 2	2.9E + 3	1.3E + 3
C°	1.7E − 1	4.0E + 0	2.4E + 2	1.1E + 2	1.2E + 2
Fe	6.0E − 2	9.0E − 1	1.6E + 1	3.9E + 1	4.5E + 1
Zn	1.1E − 2	2.0E − 1	6.8E + 0	1.6E + 1	1.0E + 1
Pb	1.0E − 2	1.8E − 1	4.9E + 0	1.3E + 1	8.0E + 0
Ni	8.1E − 4	1.2E − 2	8.8E − 1	1.9E + 0	9.8E − 1
sum[a)]	5.6E + 0	8.3E + 1	2.4E + 3	8.9E + 3	4.1E + 3
mass (gravimetric)	1.2E + 1	2.6E + 2	6.7E + 3	1.0E + 4	5.5E + 3
mass (analysed, %)	47	32	36	89	75
standard dev. (%)	170	160	51	24	10

[a)] S taken as $SO_4^=$.

The sum of the analysed elements is compared to total particulate mass inverted from the electrobalance data. The standard deviation gives the combined relative errors in the inversion of the individual chemical components in the sum.

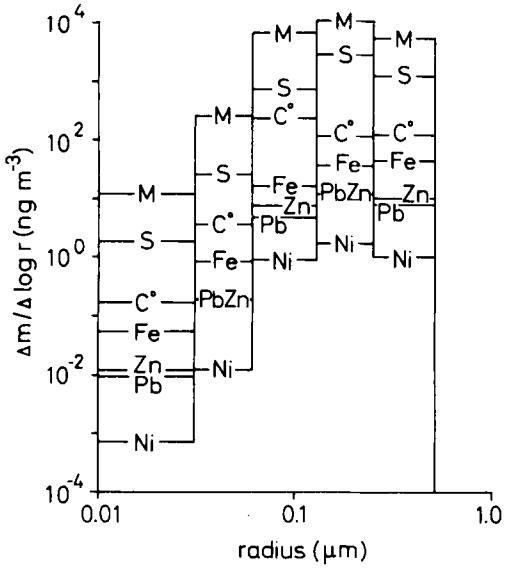

Fig. 6. Chemically resolved mass size distribution of Arctic haze at Ny-Ålesund, Spitsbergen, average of samples 4–6 (21–28 March 1983). *M* = total mass, S = non-marine sulfur, C° = elemental carbon.

6. Summary and conclusions

In March 1983, we performed a field experiment at Ny-Ålesund, Spitsbergen to study the chemical composition of Arctic haze aerosols. We derived chemically resolved submicrometric mass size distributions of 6 aerosol samples. The time resolution of our sampling procedure was about 48 h. Hence, we were not able to resolve short term air mass changes that we consider to be necessary for the identification of source regions as was possible with the results of Martinsson et al. (1984) at lower latitude. At the same time, we derived the first Arctic results on the mixing of elemental carbon and hygroscopic substances in the aerosol. The data showed an external mixture of hydrophobic soot and hygroscopic sulfur and metal components which were very similar to our corresponding urban results in Stockholm, Sweden (Covert and Heintzenberg, 1984). Admittedly, more evidence is needed before drawing general conclusions about the hygroscopic properties and external mixing of Arctic haze. A speculation about the radiative effects of Arctic on the basis of our first results would yield minimum atmospheric heating by externally mixed light absorbing particles as compared to internally mixed soot particles. Wendling et al. (1985) calculated local heating rates due to aerosols absorbing in the solar spectral range for airborne radiation and aerosol measurements during our experiment. When comparing results for instantaneous heating rates for clear and hazy conditions, they find an increase in the heating rate near surface from 0.49 (clear) to 1.25 K day^{-1} (hazy, internal mixture) and 0.28 K day^{-1} (hazy, external

mixture). These findings demonstrate the importance of the knowledge and degree of internal mixing for aerosol species when assessing the climatic effect of Arctic haze. A synopsis of size distribution and hygroscopic properties yields a very consistent picture of Arctic haze consisting of aged midlatitude aerosol particles which maintain their anthropogenic urban characteristics even after 5000 km or 5 days of transport into the Arctic.

7. Acknowledgements

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