Tellus (1981), *33*, 162–171

The chemical composition of arctic haze at Ny-Ålesund, Spitsbergen

By J. HEINTZENBERG, *Department of Meteorology*,[1] *University of Stockholm, Arrhenius Laboratory, S-106 91 STOCKHOLM, Sweden*, and H.-C. HANSSON and H. LANNEFORS, *Department of Nuclear Physics, Lund Institute of Technology, Sölvegatan 14, S-223 62 LUND, Sweden*

(Manuscript received June 13; in final form July 21, 1980)

ABSTRACT

Samples of arctic haze particles were collected at Ny-Ålesund, Spitsbergen (12° E, 79° N) during late winter 1979. The filter samples were analysed wet chemically and by particle-induced X-ray emission (PIXE). The concentrations of 17 major ions and metals were determined and compared to previous results of arctic and antarctic research. Simultaneous measurements of the aerosol size distribution were used to check the consistency of the results. Both the chemical composition and the size distribution data support the hypothesis that the arctic winter aerosol is heavily influenced by a well-aged continental aerosol originating in midlatitude anthropogenic source areas.

1. Introduction

The continuously growing anthropogenic influence on the environment causes a need for establishing baseline or background values for the different trace substances in the atmosphere. The same need exists for optical properties of the atmosphere such as visibility or turbidity since the relationship between the physico-chemical properties of the atmospheric particulates and their optical properties has not yet been clarified completely.

Reliable baseline aerosol measurements are best accomplished as far away as possible from strong source regions. In the Northern Hemisphere this leads to the central regions of the North Pacific, the North Atlantic Ocean and the polar region. The maritime regions are not well suited for ground level aerosol measurements because the local sea spray component will influence the concentrations of many elements and compounds. Thus remains the arctic region. A closer look shows that extensive atmospheric studies are justified in the Arctic not only for global baseline monitoring but also because of the specific regional character of the Arctic.

In winter, 90% of this vast area between North America and Eurasia is covered by ice and about 60% in summer. The pack ice floats like a thin film on the Arctic Ocean and causes the continental thermal character of the area. This pseudo continent has a surface albedo at about 80% but a reduction from 70 to 60% would destroy the pack ice in 8–10 years. A summer air temperature anomaly of +2 °C would cause the same destruction process in a few decades (Budyko, 1962). Global changes of climate are to be expected as a consequence of such a drastic change. Because of their scattering and absorption effects in both the solar and the terrestrial spectral regions, aerosol particles can contribute to an air temperature anomaly. In deposited form the particles can cause surface albedo changes. Imbedded in snow, absorbing particles can cause temperature changes in the snow layer.

During the winter period the arctic sink region provides a unique possibility for studying long-distance transport and transformation processes of continental aerosols emitted in Eurasian regions. On the 3000 to 5000 km long pathway from these

[1] Contribution No. 415.

0040-2826/81/020162-10$02.50/0

sources to the Arctic, the investigation of transport and transformation mechanisms are facilitated by the fact that the lower boundaries are snow- and ice-covered land and sea areas without significant gas or particle sources. The removal of atmospheric trace substances through precipitation is the most complicated aspect in long-distance transport studies. However, in the Arctic, precipitation in general is lower than in mid-latitudes and has its minimum in late winter or spring, again facilitating the study of aerosol transport. Despite these simplifying regional characteristics, only very few transport studies of the origin and the fate of the arctic aerosol have been done (Rahn et al., 1979c; Rahn, 1979b) and the results are still tentative.

A knowledge of the arctic air chemistry is needed for more quantitative modelling of trace substances in the Arctic. There have been a number of field studies mainly in Alaska (Rahn, 1971) and Greenland (Flyger et al., 1978).

Results from areas closer to the Eurasian source region (i.e. Spitsbergen, Franz-Josefs Land) have not yet been published and no serious attempt has earlier been made to combine measurements of physical (especially optical) and chemical properties of arctic haze particles. Recently a first analysis of chemical measurements in the Spitsbergen region has been presented by Larsson and Hansson (1979) on a WMO-Conference in Boulder, Co. In a field experiment conducted by one of the authors (J.H.) near Ny-Ålesund, Spitsbergen, in April–May 1979, *in-situ* measurements of the aerosol size distribution and integral optical properties were combined with the sampling of particles in two size classes for subsequent chemical analysis. Size distribution and optical properties have been reported in a previous paper (Heintzenberg, 1980). Here the results of the chemical analysis and an investigation of the air mass origin are presented. The internal consistency of the physical and chemical aerosol data is verified by comparing aerosol masses derived through the inversion of optical data with aerosol masses from chemical analysis.

2. Location and the problem of local contamination

The geographical coordinates of the sampling site Ny-Ålesund are 12° E, 79° N. Details about the station are given by Heintzenberg (1980). The risk of local contamination is the most important problem connected with the sampling of atmospheric trace substances in extremely clean remote areas. During continuous atmospheric measurements, periods of local influence can usually be clearly identified and excluded from the data evaluation by distinct level changes or increasing variance of the recorded parameters. In contrast, the sampling of aerosol under background conditions can be much more seriously affected by local contamination. Even with high-volume samplers it may take more than 1 day of sampling to get enough mass for a chemical analysis accumulated on the filters. Short periods with elevated levels through local influence can spoil several days of sampling. There is no ideal background measuring station where the absence of local contamination can be guaranteed.

In the present study a special effort was made to reduce local influence on the aerosol sampling. A General Electric condensation nuclei counter was modified to decrease the time constant of the output signal for the measuring range to about 1 sec and to lower the detection limit to about $10/cm^3$. The signal from this counter controlled the pumps of the aerosol samplers. Sampling was turned on only when the total particle concentration at the entrance of the aerosol intake was $500/cm^3$ or below. A similar arrangement has been used by Maenhaut et al. (1977). Direct emissions from the diesel oil-fired power plant of the station and from local ground and air traffic caused concentration increases by two orders of magnitude or more above the average level of about $300/cm^3$. Therefore direct local influence on our samples can safely be excluded. During our experiment, between 50 and 98% of each sampling period could be used for collecting locally uncontaminated particles.

Local contamination is not restricted to direct flow from the local sources to the measuring site. In extremely clean air masses without rapid elimination processes it is conceivable that local emissions are contaminating atmospheric measurement in aged form on indirect routes to the sampling site. Local or mesoscale circulation systems can bring aged emissions back to the source area within their residence time. We cannot prove the absence of indirect local contamination. However, during the course of the experiment a

number of observations were made which can be used as evidence:

1. The general level of the total particle concentration was in good agreement with results from other arctic measurements (Flyger et al. 1978) and with our understanding of the global background values
2. Even in periods of several days with strong winds from W–NW excluding any possibility of indirect local influence, the particle concentrations did not sink below the general level
3. While being on the background level (300/cm^3) the total particle counts usually exhibited extremely small (1–2 times the threshold value) temporal variations. During the few periods where indirect local contamination was suspected, rapid fluctuations (5–10 times the threshold value) appeared though the general concentration level remained low. Hence it was concluded that the stability of the total particle concentration could be used as an indicator for possible indirect (low level) contamination.

In Fig. 1 a section of the original Aitken nuclei recording is depicted showing both the stability of the low level particle concentrations and a period with direct local influence. The readings have to be multiplied by a factor of 2 to compensate for losses in the aerosol intake (Heintzenberg, 1980).

The combined results of particle concentration-controlled sampling and of the evaluation of the high-resolution Aitken nuclei recordings led us to the conclusion that our chemical results presented in the following sections are free from direct or indirect local contamination.

3. Experimental

3.1. *Sampling and analysis*

Sampling for chemical analysis was done by using 47 mm diameter membrane filter with 5 μm pore diameter for the total aerosol fraction and with 25 mm membrane filters for size-segregated sampling. The sequential filtration technique described by Cahill et al. (1977) employing two membrane filters in series was used. This sampler separates the aerosol particles into a coarse and a fine fraction. The flow rate used (5 l/min) yields a 50% aerodynamic cut-off radius $r_c \simeq 0.5$ μm according to Spurny and Lodge (1972) efficiency calculations, assuming a particle density of 2 g/cm^3.

The filters for the total aerosol fraction were analyzed wet chemically for nine major constituents (Heintzenberg et al., 1979). However, results from Ca^{++} are not reported because of

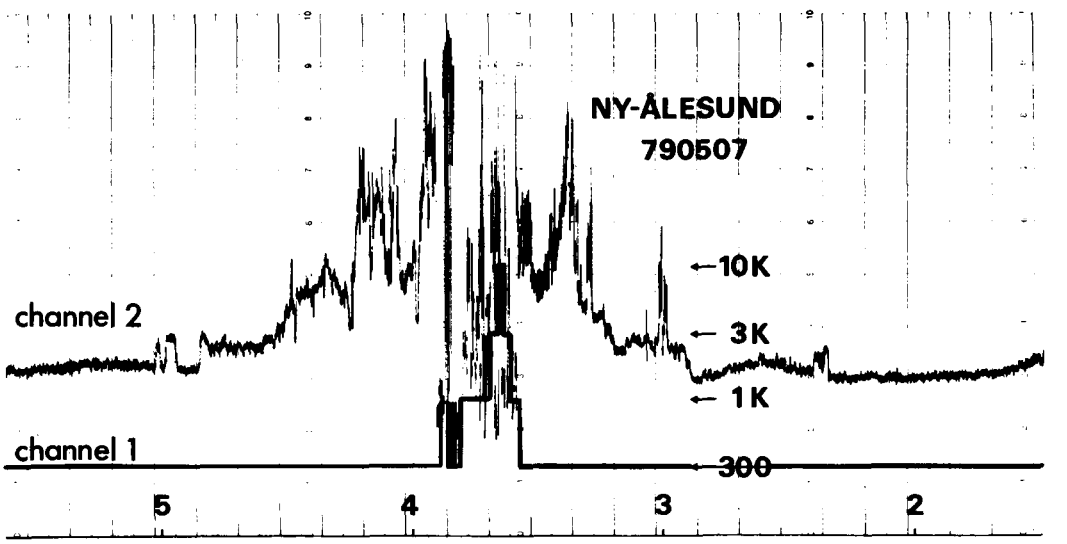

Fig. 1. Total particle concentrations at Ny-Ålesund, Svalbard, 1979-05-07. The numbers on the abscissa correspond to full hours. Channel 1 records the measuring range cm^{-3}. Channel 2 records relative particle counts in the actual range. Example: 0200 h the reading was 30% of range 300 = 90 cm^{-3}.

excessive blank values. The analysis of the size-segregated filter for sulfur and trace metals was made using particle induced X-ray emission (PIXE) (Johansson et al., 1975). The consistency of the results obtained by the two analyzing methods was checked by comparing the sulfur concentrations for our experiment. These showed satisfactory agreement, having maximum deviations of about 20%. The accuracy and precision for most of the results obtained are well below 20%. However, results with uncertainties up to 30% have been accepted in the data evaluation.

Particle size distributions and optical properties were measured with a General Electric condensation nuclei counter, a 5-channel Royco-225 optical particle counter and a 6-channel integrating nephelometer. This part of the instrumentation is described in detail in Heintzenberg (1980).

3.2. *Artifact formation in the filter sampling*

During the collection of reactive aerosols and during aerosol sampling in the presence of reactive gases, chemical reactions on the filter surfaces can introduce serious errors in the measured concentrations (Klockow et al., 1979; Pierson et al., 1980). Especially the halogens can form volatile compounds which are lost during sampling. Eriksson (1960) proposed that chloride loss occurs as a consequence of pH-lowering reactions between seasalt particles and, e.g., H_2SO_4 which causes a supersaturation of aqueous HCl in the particles relative to the atmospheric pressure of HCl vapor, resulting in volatilization of Cl as HCl. Klockow et al. (1979) have studied the loss of collected Cl and I when exposed to sulfuric acid. The relative amounts lost were influenced by filter type, relative humidity and the available amounts of hydrogen ions.

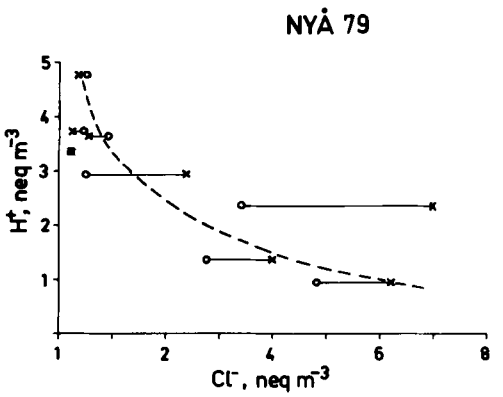

Fig. 2. Hydrogen equivalents plotted versus chlorine equivalents analyzed by wet chemically and PIXE (X), both filters added, in arctic haze samples at Ny-Ålesund, Svalbard, 1979-04–1979-05-08.

In Fig. 2 the concentrations of hydrogen ions are plotted versus chlorine for our Spitsbergen data. The chlorine concentrations appear to be inversely related to the hydrogen ion concentration. The size fractionated samples have lower losses especially in cases when the hydrogen ion concentration is low. Different artifact formation on the different filter materials used for wet chemical and PIXE analysis is one possible explanation. A second explanation is based upon the different formation mechanisms for chlorine-containing particles originating mainly from sea spray droplets and the hydrogen-containing particles of anthropogenic origin existing probably in the form of sulfuric acid or acidic sulfates. The sea spray generation mechanism will produce particles predominantly in the coarse fraction while the anthropogenic hydrogen will be found mainly in accumulation range particles (see Whitby, 1978, for definitions). Since the two ranges to a great extent will be sampled on different filters with the sequential technique, artifact formation with chlorine loss can be expected to be lower in this sampler than on the total filter sampler where all chemical species are accumulated on the same filter.

There are other possibilities of artifact formation, especially those involving SO_2 conversion. As data on gas-phase components are lacking, we must assume that there was no artifact formation involving SO_2 or HNO_3, etc.

4. Results

4.1. *Chemical composition of arctic haze samples on Spitsbergen*

Earlier measurements in the Arctic have shown very similar sulfate concentrations in northern Scandinavia, Spitsbergen and Alaska (Rahn et al., 1979c). Furthermore, these measurements show large seasonal variations, typically one order of magnitude for sulfate and vanadium (Rahn et al., 1979b). As reasons for the high winter values, they suggest the general winter circulation favoring south-north atmospheric transport to the Arctic

and the low arctic winter precipitation yielding long residence times for the arctic aerosol particles.

During our experiments at Ny-Ålesund 1979-04-19 to 1979-05-08 eight aerosol samples were collected covering time periods between 24 and 84 h. The results from the first sample were not included in the data evaluation because sampling was not yet controlled and contamination by heavy ground traffic nearby was suspected.

The results from the remaining seven samples showed a very stable absolute and relative aerosol composition. Most of the elemental concentrations varied less than 50% of their mean values, which is quite consistent with our results on the physical aerosol properties (see Table 2 in Heintzenberg, 1980). Especially the sulfate concentrations were very stable, varying less than 20% of the mean value. Chlorine concentrations exhibited the largest variability, up to one order of magnitude. However, the true variation cannot be separated from variations caused by the artifact discussed earlier. Vanadium is commonly used as a tracer for anthropogenic influence (combustion of heavy oil) in atmospheric aerosols. Unfortunately, we were not able to detect V in our own samples and refer to late winter measurements 1973–1974 at Ny-Ålesund (NILU, 1977). Our grand average results are compared with averages from other locations in Table 1. The concentrations at the rural site are in general on the same level or slightly higher than those on Spitsbergen. Exceptions are iron, zinc and lead concentrations being 3–5 times higher at the rural site.

Concentration levels of urban aerosol in Copenhagen, Denmark (Flyger et al., 1976) are one to two orders of magnitude higher than on Spitsbergen, with the exception of sulfur being only 4–5 times higher in the urban aerosol. In the urban source area, a large fraction of the sulfur in the atmosphere is in the primary form of SO_2, while in the arctic sink region a large part of the SO_2 can be expected to be transformed into particulate sulfate. Hence the relatively high particulate sulfur values on Spitsbergen do not necessarily reflect high pollution levels.

The South Pole is located near the center of the snow-covered Antarctic continent. Though it is much more isolated from tropospheric sources than any of the other remote locations considered, a comparison is somewhat complicated because of its height and the possibility of subsiding stratospheric air carrying sulfate particles to the surface.

Table 1. *Average chemical composition of atmospheric aerosol samples at different locations. Concentrations are given in ng m^{-3}*

Element	Urban Copenhagen*	Rural Velen†	Remote Summer South Pole‡	Remote Ny-Ålesund Late winter Spitsbergen§	Remote Summer N. Greenland‖
Na	—	—	3.3	130	—
Mg	—	—	0.72	51	—
Si	3700	—	—	110	43
S	3100	—	49	690	27
Cl	620	140	2.6	(96)	10
K	460	56	0.68	32	3.8
Ca	1600	58	0.49	73	9.1
Ti	70	4.1	0.10	<5.2	0.85
V	29	2.0	0.0013	0.2¶	0.12
Cr	7.5	3.0	<0.04	2.6	0.09
Mn	37	4.7	0.013	1.5	0.21
Fe	1200	61	0.62	64	6.4
Ni	11	1.3	—	0.7	0.08
Cu	25	2.0	0.029	<1.9	—
Zn	360	17	0.033	3.2	0.18
Pb	1700	16	0.076	<4.9	0.20

* Flyger et al. (1976). † Lannefors et al. (1979). ‡ Maenhaut et al. (1977). § Present work. ‖ Flyger et al. (1978). ¶ NILU (1977).

Hence, excluding S from the comparison yields values between 10 (Fe) and 150 (V) for the ratio north polar winter concentration/south polar austral summer concentrations and values between 2 (Cr) and 90 (V) for the ratio north polar summer concentrations/south polar summer concentrations. The extremely low vanadium concentrations in the south polar atmosphere indicate how effective the isolation of the location from anthropogenic sources is. Crustal weathering on the other hand seems to be a particle source common to both polar regions, which is somewhat surprising considering the extent of snow and ice coverage on the land masses. Fe comprises about 6% of the average crustal material and is found to be the dominating species of the metal components considered in the polar air samples. The lack of seasonal variation, which is confirmed by the agreement with mean annual concentrations found on Novaya Zemlya (Egorov et al., 1970), also indicates a local (crustal) source for Fe. Because of the lack of local crustal references, we refrain from a more detailed evaluation of the crustal component in our samples.

In earlier studies the seasonal variation of sulfur and vanadium in the arctic aerosol was reported. The summer results by Flyger et al. (1978) in northern Greenland (82.15° N, 29.88° W) can be used to give a rough idea about winter–summer variations for a large number of elements. The late winter Spitsbergen data are typically one order of magnitude higher than the summer Greenland data. Especially the known anthropogenically influenced elements such as sulfur, zinc and lead have 20–30 times lower concentrations in summer. These drastic seasonal variations are consistent with the coupling-decoupling hypothesis proposed by Rahn et al. (1977) and with a first analysis of results from the Norwegian arctic measuring programme (Larsson et al., 1979). The central idea is that the Arctic is cut off or decoupled from the midlatitude source regions by the polar front in summer. In winter, on the other hand, the polar front lies just south of the midlatitude source areas.

This means that in winter transport to the Arctic is facilitated at the same time as atmospheric elimination processes along the pathways are less effective than in summer.

4.2. *Ionic balance and molecular composition*

As a means of checking the completeness of the chemical analysis, cation-anion balances were calculated for all samples. The results are listed on an equivalent basis in Table 2. Because of high blank values in the wet chemical analysis or because of the Cl-losses discussed in section 3, PIXE results have been used for the elements Cl, K and Ca. The sum of all analysed metals (M^+) also is taken from the PIXE analysis.

The fact that we always found an excess of cations might be attributed to phosphates and silicates missing in our analysis. As a relative measure of the completeness of the chemical analysis we calculated the parameter $\Delta = (\Sigma^+ + H^+ - \Sigma^-)/(\Sigma^+ + H^+ + \Sigma^-)$. The sum of all cations minus the anions yields the number of unbalanced charges which is compared to the total number of charges accounted for by our analysis. Δ is listed in the last column of Table 2. Our analysis yielded charge balance within about 10%.

The two charge-dominating ions are SO_4^{2-} and H^+. If subtracting the sum of all cations from the

Table 2. *Ionic balance on equivalent basis in aerosol samples taken at Ny-Ålesund, Svalbard, 1979-04-19–1979-05-08. M^+ stands for the sum of metal cations. $\Delta = (\Sigma^+ + H^+ - \Sigma^-)/(\Sigma^+ + H^+ + \Sigma^-)$. The values are given in neq/m³.*

Sample	SO_4^{2-}	NO_3^-	Cl^-	Na^+	K^+	Mg^{2+}	NH_4^+	M^+	Σ^-	Σ^+	H^+	Δ %
1	31.3	0.4	6.3	9.1	1.9	5.0	7.5	19.4	38.0	42.9	9.5	+16
2	43.1	2.0	0.6	1.8	0.2	1.6	16.2	2.1	45.7	21.9	36.9	+13
3	61.9	0.9	0.4	1.3	0.5	4.0	14.1	2.9	63.2	22.8	47.8	+6
4	41.9	0.3	0.2	1.9	0.7	2.1	9.2	3.9	42.4	17.8	33.5	+9
5	46.3	0.4	0.3	2.3	1.0	2.8	10.6	3.9	47.0	20.6	37.3	+10
6	33.1	0.3	4.0	9.1	1.0	5.4	6.4	5.4	37.4	27.3	13.8	+5
7	52.5	0.1	2.4	6.5	1.3	6.3	11.2	5.2	55.0	30.5	29.5	+4
8	45.6	0.6	7.1	11.7	0.9	6.7	8.6	4.0	53.3	31.9	23.5	+2

sulfate equivalents yields a residue of SO_4^{2-}, these ions must have protons as partners yielding sulfuric acid. Doing this in Table 2 indeed gives substantial amounts of sulfuric acid for all samples except number one which we classify as locally contaminated (mainly by metals).

The acidity of the arctic aerosol also can be discussed by just considering the measured pH-values. On the average, the H^+ concentrations were 32 ng/m³. In section 5.3 a total suspended dry mass of 4 μg/m³ is derived. There was no information on relative humidity available during the experiment. However, assuming the amount of water in the airborne aerosol particles to have the same magnitude as the dry particles, we derive a pH of about 2 for the airborne material. Hence our chemical analysis indicates that the arctic aerosol is very acidic, probably containing substantial amounts of sulfuric acid and acid sulfates.

5. Discussion

5.1. *Air mass analysis*

A trajectory analysis based upon isobaric 850 mbar 96 h back trajectories calculated by the Norwegian Institute of Meteorology did not yield conclusive results about the air mass origin. There are three reasons for this: (1) Ny-Ålesund lies in the N.W. corner of the trajectory grid system, (2) the meteorological information for the trajectory calculations is very limited in the arctic region, (3) Because of the small pressure differences in the Spitsbergen area only weak air movements occurred during the whole experiment. However, the finding that (as far as trajectories could be calculated) the air masses reaching the station had passed mainly over ice covered areas of the Arctic Ocean is noteworthy. Only during parts of samples 6 and 7 had the air passed over the Norwegian Sea and the northernmost parts of Scandinavia.

5.2. *Chemical composition as a function of particle size*

The average chemical composition of the haze particles is calculated in two size fractions divided at $r_c = 0.5$ μm (see Table 3). The three major sources contributing to the arctic aerosol are the ocean, the earth's crust and anthropogenic sources. The first two produce mainly aerosols in the coarse particle range while the third is dominating the fine particle and accumulation modes. The marine source has a rather small influence because the air has passed mainly over ice-covered sea areas. Part of the $SO_4^{=}$, Cl^-, Na^+ and to lesser extent Mg^{++} and Ca^{++} are probably of marine origin. According to the generation mechanism their major fraction lies in the coarse particle range. Typical soil derived elements are Si, Ca, Ti, Fe (and to some extent Mg, K, Cr and Mn). They also are found mainly in the coarse particle mode in accordance with their production process. SO_4, NH_4, NO_3, Cu, Zn and Pb are associated predominantly with submicron aerosols in urban areas.

Table 3. *Average size segregated chemical composition of arctic haze samples at Ny-Ålesund, Svalbard, 1979-04-19–1979-05-08. Total suspended mass in accumulation range and coarse particle range are compared to results from inversion of optical data*

Element or ion	Average concentrations, ng/m³ Accumulation range	Coarse particle range
H	32*	—
NH_4	153*	—
NO_3	39*	—
Na	—	33[1)]
Mg	—	49[1)]
Si	41	68
SO_4	1850	220
Cl	(11)	(85)
K	15	17
Ca	21	52
Ti	<2.0	3.2
Cr	1.6	1.0
Mn	0.75	0.75
Fe	26	37
Cu	1.6	<0.32
Zn	2.6	0.58
Pb	3.0	<1·9
Chemical analysis (excl. sample 1)	Σ2200	Σ650
Inversion of optical data (grand average)	Σ3900	Σ760

* Results of total aerosol samples, assumed size segregation.

5.3. *Comparison between chemical and optical analysis*

The relationship between the light scattering coefficient σ_{sp} and submicron mass concentration m_{acc} or submicron sulfate concentration $(SO_4^{2-})_{acc}$

has been used as an index of the role of submicron particles and especially sulfates in tropospheric optics. The ratios σ_{sp}/m_{acc} and $\sigma_{sp}/(SO_4^{2-})_{acc}$ have been determined for continental aerosols in many different locations (Waggoner et al., 1976; Waggoner et al., 1980). For the grand average results we find $\sigma_{sp}/m_{acc} = 3.9$ m^2 g^{-1} which is in good agreement with previously reported data for industrial regions. The light scattering sulfate ratio at Ny-Ålesund is 8.3 m^2 g^{-1} which is near 10 m^2 g^{-1} reported by Waggoner et al. (1976) for St. Louis, Missouri.

The size distribution inverted from our optical measurements (Heintzenberg, 1980) can be written as mass size distribution assuming a density of 2 g cm^{-3} for the aerosol particles. This inverted mass distribution from the grand average of the optical data has been plotted in Fig. 3. In general inverted size distributions depend on the refractive index of the aerosol particles. The results of the chemical analysis can be explained assuming acid sulfate particles dominating arctic haze (see Tables 1–3). Therefore in the inversion we allowed the refractive index of the particles to vary between 1.36–0i (sulfuric acid) and 1.53–0i (ammonium sulfate). The corresponding variations in the inverted size distribution are marked as error bars in Fig. 3. Drawn as a histogram are the inversion results for the refractive index 1.5–0.005i which we consider to be a good approximation for background aerosol particles. The mass size distribution was integrated over the size ranges of our two-stage sampler. The results are compared with the mass results from chemical analysis in Table 3.

Both chemical and optical analyses yield 3–5 times more mass in the accumulation range than in the coarse particle range. The total suspended mass from chemical analysis is 3 μg/m^3 compared to 5 μg/m^3 from the inversion. The chemical status of the elements analysed by PIXE have not been taken into the calculations. Presumably many of the metals exist as oxides. A number of main components, i.e. C, Al, organics have not been determined. Adding those components to the mass chemically determinable would bring the optical and chemical total suspended mass concentrations into closer agreement. Furthermore, considering the fact that optical and chemical analyses are looking at completely different particle properties and both involve a number of assumptions, the agreement is quite satisfactory.

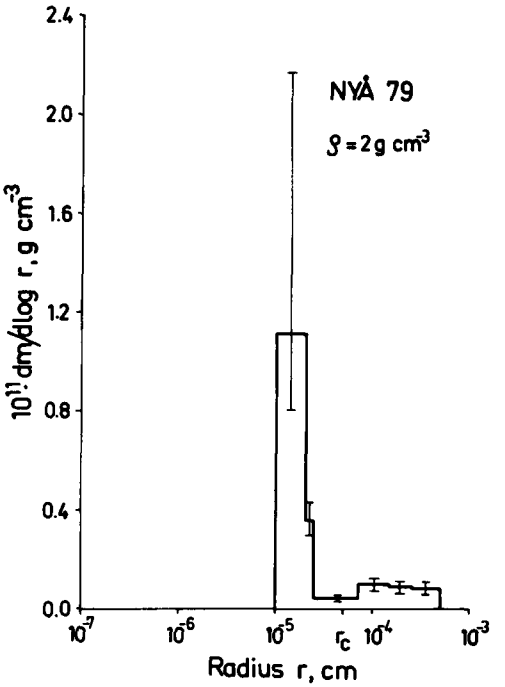

Fig. 3. Grand average mass size distribution inverted from optical measurements at Ny-Ålesund, Svalbard, 1979-04-19–1979-05-08. The density was assumed to be 2 g cm^{-3}. The results for $m = 1.5$–0.05i are drawn as a histogram. The error bars show the variation of the results when allowing refractive index variations between $m = 1.36$–0i (sulfuric acid) and $m = 1.53$–0i (ammonium sulfate). r_c is the 50% cut off radius of the two-stage sampler for chemical analysis.

6. Conclusions

All evidence of previous studies of the composition and the origin of the arctic aerosol (see the review by Rahn, 1979c) suggests the anthropogenic pollution component dominating the winter aerosol in the Arctic. In this study, the combination of chemical composition, optical properties and size distribution data independently support this hypothesis. The mass size distribution determined by physical measurements has its sharp maximum in the accumulation range, indicating the presence of a well-aged aerosol. The results of the size fractionated chemical measurements show that S, Cu, Zu and Pb are found mainly in submicron particles. Long-range transport from anthropogenic, midlatitude source areas seems to be the only possible cause for these findings.

7. Acknowledgements

We thank the Norwegian Polar Institute for the help with the field experiment at Ny-Ålesund, Spitsbergen. The Arctic Expedition Ymer-80 is gratefully acknowledged for financial support. We acknowledge the Norwegian Meteorological Institute for calculating the back trajectories.

REFERENCES

Budyko, M. I. 1962. Polar ice and climate. Izvestiya Akademiya Nauk SSSR, Seriya Geographicheskaya No. 6, quoted in Fletcher J. O. 1965. The heat budget of the arctic basin and its relation to climate. Rand Report R-444-PR.

Cahill, T. A., Ashbaugh, L. L., Barone, J. B., Eldred, R. A., Feeney, P. J., Floccini, R. G., Goodart, C., Shadoan, D. J. and Wolfe, G. W. 1977. Analysis of respirable fractions in atmospheric particulates via sequential filtration. *J. Air Poll. Contr. Assoc. 27*, 675–678.

Egorov, V. V., Zhigalovskaya, T. N. and Malakhov, S. G. 1970. Microelement content of surface air above the continent and the ocean. *J. Geophys. Res. 75*, 3650–3656.

Eriksson, E. 1960. The yearly circulation of chloride and sulfur in nature; meteorological, geochemical and pedological implications. Part II. *Tellus 12*, 63–109.

Flyger, H. and Heidam, N. Z. 1978. Ground level mesurements of the summer tropospheric aerosol in northern Greenland. *J. Aerosol Sci. 9*, 157–168.

Flyger, H., Palmgren-Jensen, F. and Kemp, K. 1976. Air pollution in Copenhagen. Part I: Element analysis and size distribution of aerosols. Risø Report No. 338, Danish Atomic Energy Commission, Risø, Roskilde,, Denmark.

Heintzenberg, J., Trägårdh, C., Alm, G., Boström, R., Granat, L., Hansson, H-Ch., Holm, B., Moritz, S., Richter, A. and Söderlund, R. 1979. Physical and chemical properties of aerosols under varying European influence. Report AC-48, Department of Meteorology, University of Stockholm. (Available from the author.)

Heintzenberg, J. 1980. Particle size distribution and optical properties of arctic haze. *Tellus 32*, 251–260.

Johansson, T. B., van Grieken, R. E., Nelson, J. W. and Winchester, J. W. 1975. Elemental trace analysis of small samples by proton induced X-ray emission. *Anal. Chem. 47*, 855–860.

Klockow, D., Jablonski, B. and Niessner, R. 1979. Possible artifacts in filter sampling of atmospheric sulfuric acid and acidic sulphates. *Atm. Env. 13*, 1665–1676.

Lannefors, J., Hannson, H.-Ch., Akselsson, R., Bohgard, M., Granat, L., Johansson, T. B., Rudell, B. and Johansson, S. A. E. 1979. Aerosolsammansättning i ett renluftsområde. Slutrapport för SNV 7-147. (In Swedish). Dept. of Nuclear Physics, Lund Institute of Technology, Lund, Sweden.

Larsson, S. and Hanssen, J. E. 1979. Annual variation and origin of aerosol components in the Norwegian arctic-subarctic region. Paper presented at the WMO Technical Conference Relative to Climate, August 20–24, 1979, Boulder, CO, U.S.A.

Maenhaut, W. and Zoller, W. H. 1977. Determination of the chemical composition of the South Pole aerosol by neutron activation analysis. *J. Radioanal. Chem. 37*, 637–650.

NILU, 1977. Annual Report 1977, Norwegian Institute for Air Research, Lillestrøm, Norway.

Pierson, W. R., Brachaczek, W. W., Korniski, T. J., Truex, T. J. and Butler, J. W. 1980. Artifact formation of sulfate, nitrate, and hydrogen ion on backup filter: Allegheny Mountain Experiment. *J. Air Poll. Contr. Assoc. 30*, 30–34.

Rahn, K. A. 1971. Sources of trace elements in aerosols—An approach to clean air. Ph. D. Thesis, University of Michigan, Department of Atmospheric and Oceanic Sciences.

Rahn, K. A., Borys, R. D. and Shaw, G. E. 1977. Particulate air pollution in the Arctic: Large-scale occurrence and meteorological controls. Proceedings of the 9th International Conference on Atmospheric Aerosols, Condensation and Ice Nuclei, Galway, Ireland, 21–27.

Rahn, K. A. and McCaffrey, R. J. 1979a. On the origin and transport of the winter Arctic aerosol. Paper presented at the Conference on Aerosols: Anthropogenic and Natural-Sources and Transport, January 1–12, New York. To be published in the Annals of the New York Academy of Sciences.

Rahn, K. A. 1979b. The Eurasian sources of arctic aerosol. Norwegian Institute for Air Research. Oppdragsrapport No. 34/79.

Rahn, K. A. and McCaffrey, R. J. 1979c. Long-range transport of pollution aerosol to the Arctic: a problem without boarders. Paper presented at the WMO-Symposium on Long-Range Transport of Pollutants and its Relation to General Circulation Including Stratospheric/Tropospheric Exchange Processes. Sofia, Bulgaria, 1–5 Oct., 1979.

Spurney, K. R. and Lodge Jr, J. P. 1972. Collection Efficiency tables for membrane filters used in the sampling and analysis of aerosols and hydrosols, NCAR Technical Note, Vols. I–III.

Waggoner, A. P., Vanderpol, A. J., Charlson, R. J., Larsson, S., Granat, L. and Trägårdh, C. 1976. Sulphate-light scattering ratio as an index of the role of sulphur in tropospheric optics. *Nature 261*, 120–122.

Waggoner, A. P. and Weiss, R. E. 1980. Relationship of light scattering and fine particle mass concentration. *Atm. Env.* (in press).

Whitby, K. T. 1978. The physical characteristics of sulfur aerosols. *Atm. Env. 12*, 135–159.

ХИМИЧЕСКИЙ СОСТАВ АРКТИЧЕСКОЙ ДЫМКИ В НЮ-ЭЛЕЗУНД, ШПИТЦБЕРГЕН

В конце зимы 1979 г. в Ню-Элезунде на Шпитцбергене (12°в.д., 79°с.ш.) были собраны пробы частиц арктической дымки. Пробы с фильтров анализировались химически (в растворе) и рентгено-флюоресцентным методом. Были определены концентрации 17 основных ионов и атомов металлов, которые сравниваются с результатами предыдущих арктических и антарктических исследований. Одновременные измерения распределения частиц аэрозоля по размерам использовались для проверки согласованности полученных результатов. Как химический состав, так и распределение по размерам поддерживают гипотезу, что зимний арктический аэрозоль испытывает большое влияние долгоживущего континентального аэрозоля, возникающего в областях антропогенных источников средних широт.