Tellus (1983), *35B*, 40–54

A comprehensive study of physical and chemical parameters of the Arctic summer aerosol; results from the Swedish expedition *Ymer-80*

By HANS LANNEFORS[1], JOST HEINTZENBERG[2] and HANS-CHRISTEN HANSSON[1],
[1]*Department of Nuclear Physics, Lund Institute of Technology, Sölvegatan 14, S-223 62 Lund, Sweden,*
[2]*Department of Meteorology, University of Stockholm, Arrhenius laboratory, S-106 91 Stockholm, Sweden**

(Manuscript received April 18; in final form June 4, 1982)

ABSTRACT

The Swedish icebreaker expedition *Ymer-80* exploring the Norwegian part of the Arctic during the summer of 1980 offered unique opportunities for a coordinated atmospheric research program. Chemical and physical properties of the Arctic aerosol were studied using different kinds of samplers and continuous monitors connected to a common air inlet. Local contamination such as emissions from the ship and its helicopters were avoided using a condensation nucleus counter to control the sampling. Emissions from Arctic settlements were found to contribute only condensation nuclei and to have no significant influence on the aerosol mass and chemical composition. Arctic summer grand average levels, especially those of possible anthropogenic components i.e. soot, non-marine S, Ni, Cu, Zn and Pb, were lower by one order of magnitude or more than late winter levels as found on Spitsbergen, but on the same level as summer values in Northern Greenland. Most of the components varied over 1 to 2 orders of magnitude during the expedition. Sampling periods mainly influenced by sea spray were characterized by high TSP and chlorine levels. A few cases, possibly influenced by long-range transport from the European continent, were characterized by high σ_{sp}, soot, sulphur and heavy metal concentrations. The Arctic background aerosol composition was found to be determined by the relative strength of active source and sink mechanisms in combination with the sampling time resolution.

1. Introduction

The influence of midlatitudinal anthropogenic sources on the Arctic aerosol have been found to be strong during the winter period (Rahn and Shaw, 1978; Rahn and McCaffrey, 1980). According to Rahn's coupling-decoupling hypothesis, the Arctic region is coupled to the Eurasian continent during the winter (i.e. the meteorological conditions are more favourable for air mass transport from midlatitudinal large source regions into the Arctic). A low winter precipitation increases the atmospheric residence times. This enables air pollutants transported to the Arctic region to accumulate there. According to Rahn's hypothesis, the Arctic is decoupled during the summer when the precipitation is also somewhat higher, which implies lower air pollution concentrations. In fact, winter values of a number of air pollution indicators (e.g. turbidity, non-marine sulphur, manganese and vanadium etc.) as measured in Northern Alaska are an order of magnitude or more above summer values. Measurements of physical and chemical properties of airborne particulate matter during the summer in Northern Greenland (Flyger and Heidam, 1978) also indicate very low concentrations compared with those measured in late winter on Spitsbergen (Heintzenberg, 1980; Heintzenberg et al., 1981).

The Arctic summer air has the lowest air pollution levels. Because of the potential influence

* Contribution No. 458.

0280-6509/83/010040-15$02.50/0

on the energy balance it is interesting to study the chemical composition of this atmosphere. It is also of great importance to obtain reliable measurements of as many components as possible, especially those linked to human activity, before oil drilling activities start in these areas. Such activities could drastically increase the emissions in the Arctic shelf regions. An example of the rapid change in the Arctic during recent decades is that the population north of the Arctic circle tripled between 1940 and 1975 (CIA, 1978).

To the memory of A. E. Nordenskiölds discovery of the north east passage in 1878–79, a scientific expedition, using the Swedish icebreaker *Ymer* as research vessel, was undertaken in the summer of 1980. One of the larger efforts was the atmospheric (chemistry and physics) program which included several research groups from Sweden, Western Germany and the U.S. Through extensive measurements, the chemical and physical properties of the Arctic summer aerosol (and trace gases) were to be determined, leading to information about possible sources and sink mechanisms.

2. Experimental

The first leg of the expedition operated from July 5 to August 4 in the area between Spitsbergen and Franz Joseph land as shown in Fig. 1. The second leg covered the area between Northern Greenland and Franz Joseph land from August 10 to September 22, 1980 (see Fig. 1). The atmospheric program was placed on the third deck 16 m above sea level in 2 containers, one for pumps and the other for the sampling equipment (see Fig. 2). The air intake consisted of a 5 m long vertical 15 cm diameter polythene pipe with a hood on top to prevent droplets and snow flakes from entering. The pipe was bent 90° in order that it should enter the sampling container horizontally from the front. The design and positioning of the pipe was based on smoke-generation studies of the turbulence on the forward side of the bridge. The risk of contamination from resuspended material deposited on the fore deck was thus minimized and the influence from sea spray created at the bow reduced.

The samplers were mounted along the pipe inside

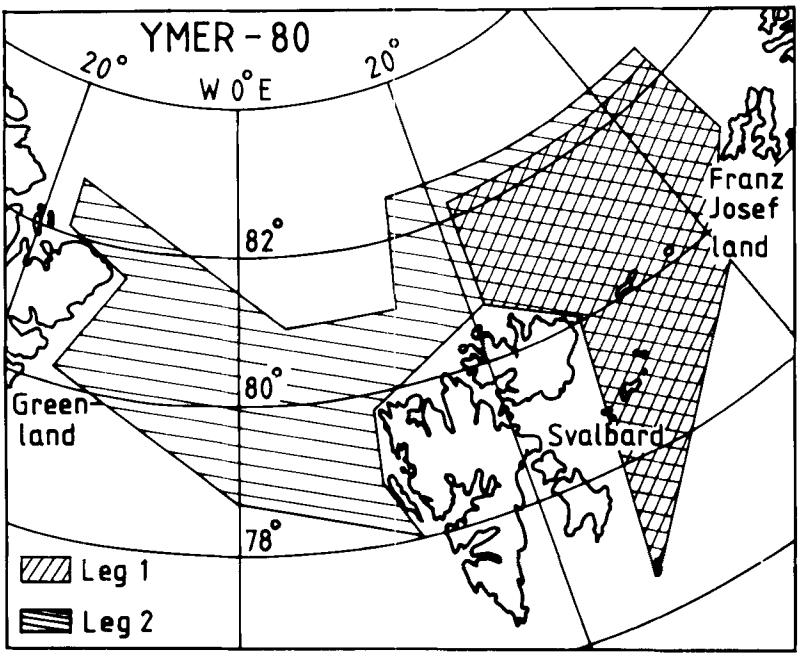

Fig. 1. Working areas during the 2 legs of the expedition.

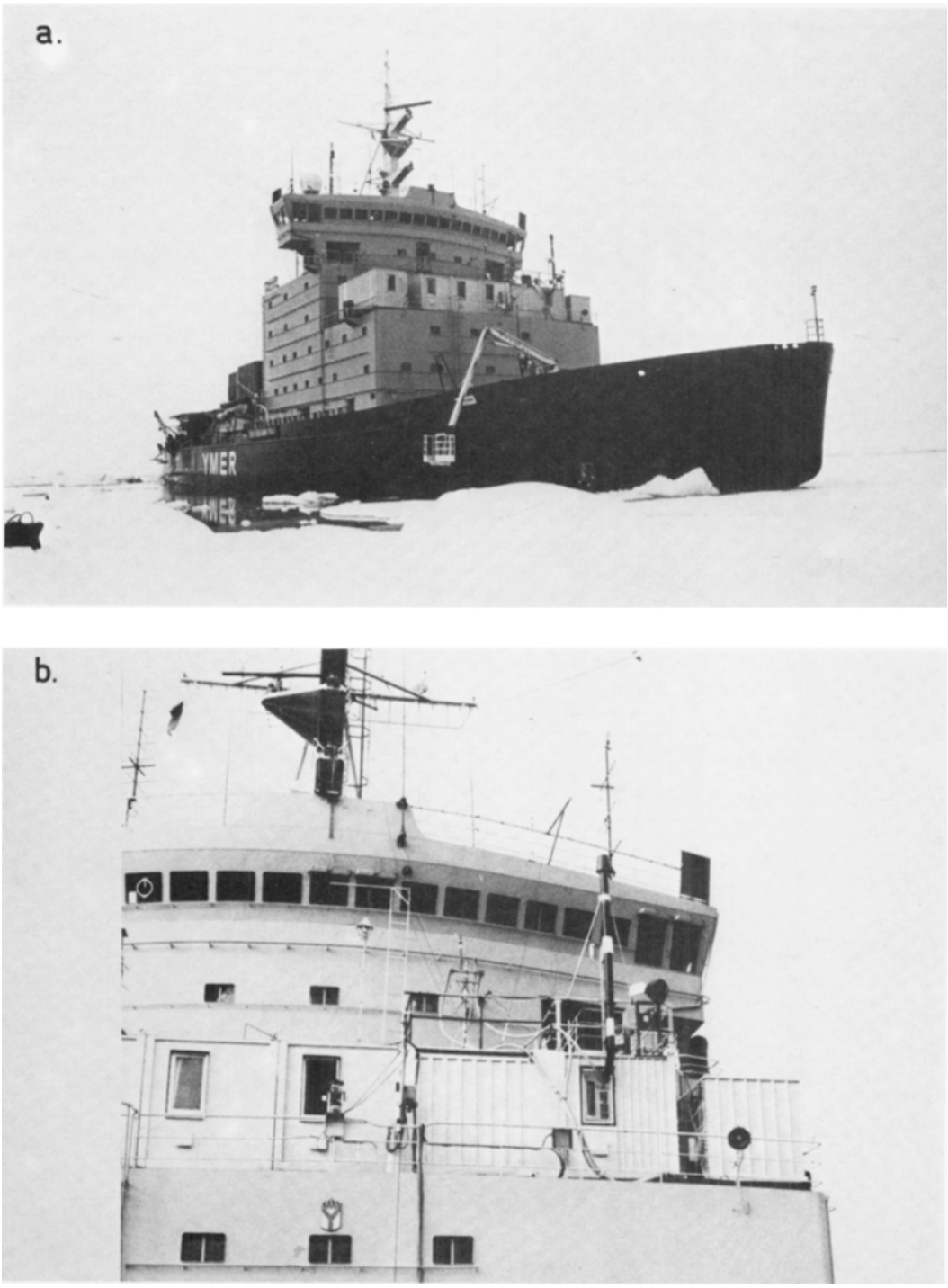

Fig. 2. (a) Photograph of *Ymer* in the Arctic ice. (b) Close-up of pump container (first from right) and sampling container (second from right) with the black inlet pipe.

the container as shown in Fig. 3. Isokinetic sampling using specially designed plexiglass inlets was employed for those samplers where the coarse fraction (particles >2 μm) was of interest. Table 1 contains a list of the aerosol samplers connected to the pipe.

Emissions from the icebreaker's diesel electric engine and activities related to the other scientific programs such as helicopter search for polar bears and leads in the packice, represented strong sources of contamination. In order to avoid contamination, the sampling was controlled by a *TSI 3020* condensation nuclei counter (CNC) which turned off all samplers when a preset concentration level was exceeded. A fast response control signal was taken directly from the photodiode to detect stack emissions while a slow response signal from the sampling output was used to indicate changes in the general level of condensation nuclei (CN) concentration. After each switch-off a 5 min delay was added to clean the whole sampling system. The CN concentration is a very sensitive indicator of recent emissions of combustion aerosol where the particles have not yet coagulated into the accumulation mode (particles in the 0.05–2 μm diameter range). The composition of the smoke emitted from the diesel engines was measured at the top of the stack using total filters. The problem of possible local indirect contamination is discussed later.

3. Results

The time series of a number of aerosol parameters measured during the *Ymer-80* expedition is shown in Fig. 4. The length of the measurement periods depends on both the prevailing aerosol concentrations and the sampling efficiency, i.e. the ratio of actual sampling time to total length of the measurement period. The duration of the measurement periods ranged from 8 h to a week and the sampling efficiency from 30 to

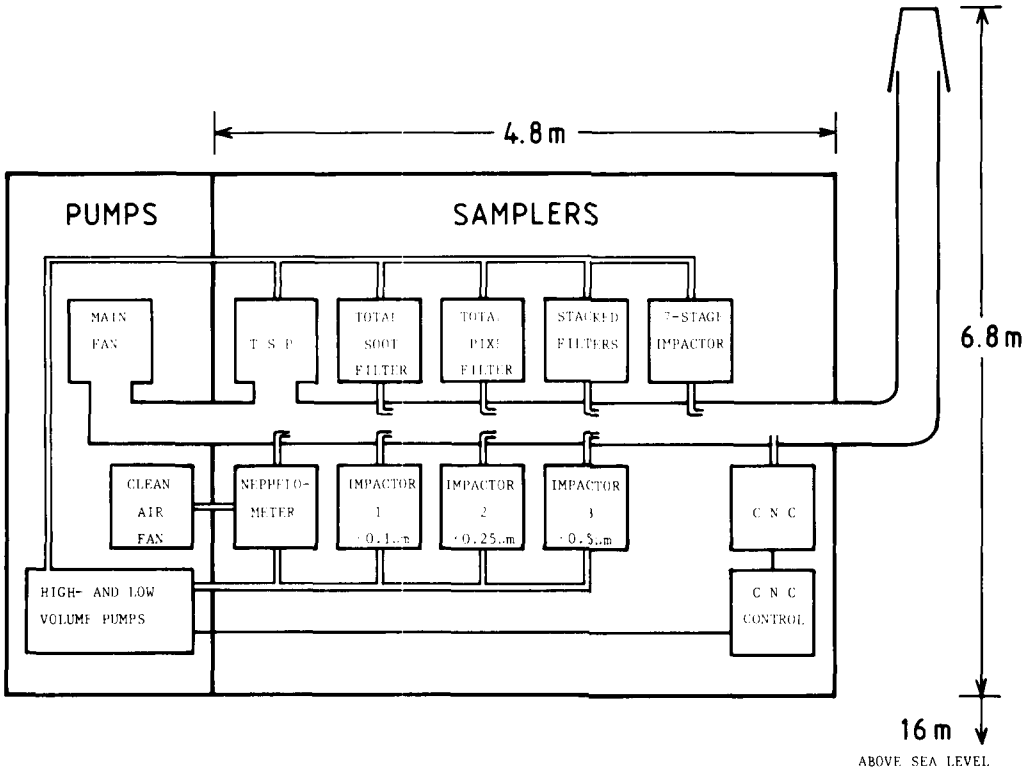

Fig. 3. Schematic figure of the air intake and samplers.

Table 1. *Characteristics of samplers*

Instrument	Sampling media	Measured aerosol parameter	Diameter range	Analytical technique	Reference
High volume sampler	142 mm ∅ ZEFLUOR 2 μm pore size	Total suspended particulate mass	$< \sim 20$ μm	Gravimetric	
Total soot sampler	47 mm ∅ MICROSORBAN filter	Total mass of soot, light absorption coefficient	$< \sim 20$ μm	ISP (Integrating sphere photometer)	Heintzenberg (1982)
Total PIXE sampler	25 mm ∅ ZEFLUOR 2 μm pore size	S, Cl, metals	$< \sim 20$ μm	PIXE (Particle Induced X-ray Emission)	Johansson et al. (1975)
Stacked filters (two-stage)	25 mm ∅ NUCLEPORE 8 μm pore size	S, Cl, metals	>2–3 μm	PIXE	Cahill et al. (1979)
	13 mm ∅ NUCLEPORE 0.4 μm pore size	S, Cl, metals	<2–3 μm	PIXE	
7-stage Battelle cascade impactor	Polystyrene with evaporated paraffin 25 mm ∅ NUCLEPORE 0.4 μm pore size	S, Cl, metals	>8 μm, 4–8 μm 2–4 μm, 1–2 μm 0.5–1 μm, 0.25–0.5 μm <0.25 μm	PIXE	Mitchell and Pilcher (1959)
Integrating nephelometer		Light scattering coefficient at 0.55 μm wavelength	mainly 0.2–1 μm	Photon counting	Charlson et al. (1974)
Winkler impactors	47 mm ∅ MICROSORBAN 15 mm ∅ ZEFLUOR 2 μm pore size	Mass of soot, *S*	<0.2 μm <0.5 μm <1 μm	ISP and PIXE	
Condensation nuclei counter, TSI 3020		Particle concentration per cm^3	0.004–0.2 μm	Optical counting of single droplets	Bricard et al. (1976)

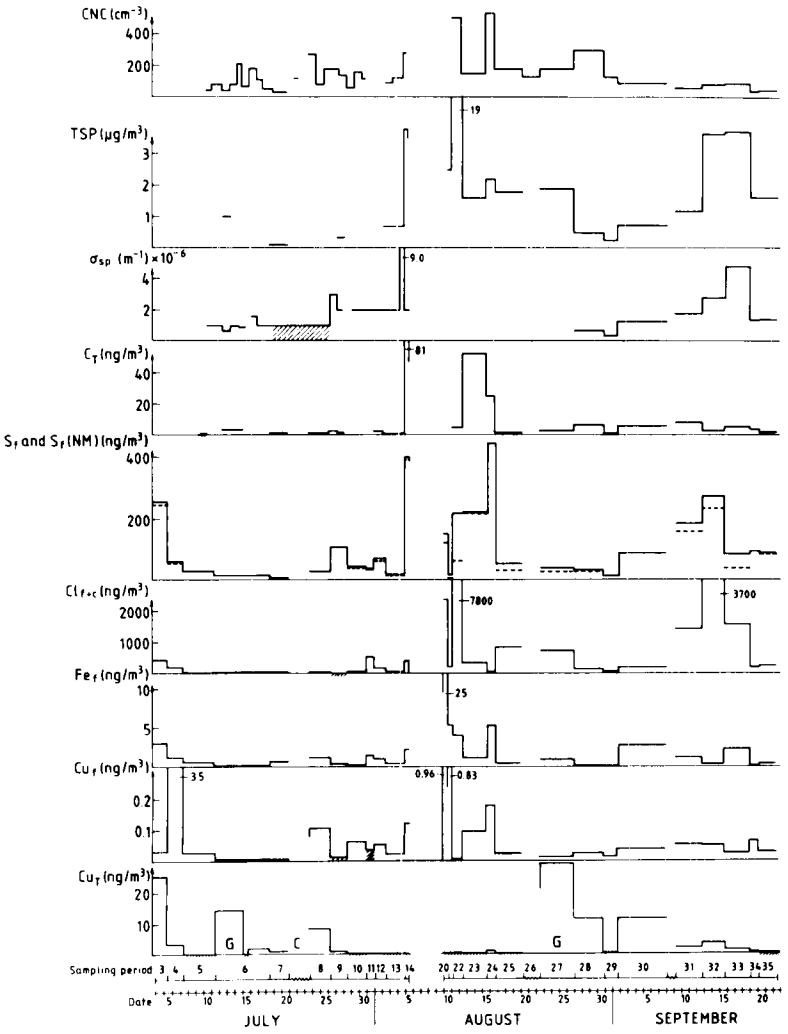

Fig. 4. Concentration versus time for 9 of the measured aerosol parameters. TSP denotes total suspended particulate matter, σ_{sp} the light scattering coefficient and C total elemental (graphitic) carbon. Index T indicates total fraction. f indicates accumulation mode, c indicates coarse mode and NM means non-marine component (marked with broken lines ---- in case of S). ///// indicates detection limit. In the Cu_T time series, G shows incidents of garbage burning and C cleaning of the inlet pipe.

almost 100%. The time constant of the variations appears to be of shorter duration than the measurement periods since often no trends can be resolved in the time series. Therefore it is probable that larger variations should be exhibited if the time resolution of the measurements were increased. In the present results, variations of up to two orders of magnitude were found for many of the parameters, which is far larger than the errors in sampling and analysis, being typically of the order of 20% or less.

Time weighted averages have been calculated and are shown in Table 2. Samples 3, 4, 14, 20, 21 and 23 were excluded from the grand average and the monthly averages since they were either taken outside the working area shown in Fig. 1 or were

Table 2. *Time weighted monthly and grand averages compared to measurements at other remote locations; concentration averages of samples representing seaspray, long-range transport and background are also listed*

		[1]	[2]	[3]		Ymer-80			Sample numbers		
Parameter	Unit	Great Smokey Mts	Ny-Ålesund Spitsbergen	Northeast Greenland	Grand average	July	August	September	22 + 33 Seaspray	14 + 24 Long-range transport	7 + 29 Arctic background
CN	cm^{-3}		380	200‡	130	100	240	76	270	390	80
TSP	$\mu g \cdot m^{-3}$	30	4.7		1.9	0.39*	3.1	1.8	11	3.0	0.17
σ_{sp}	$10^{-6}\ m^{-1}$		15		1.5	1.1	1.6*	1.9	4.8$	9.0§	0.68
C_T	$ng \cdot m^{-3}$	1100	70		4.5	1.9	5.7	4.5	5.5	53	1.3
S_f	$ng \cdot m^{-3}$	3700	620	27	99	42	100	130	150	300	13
$S_{f_{NM}}$	$ng \cdot m^{-3}$	3700	620	26	82	41	76	110	53	300	13
Cl_{f+c}	$ng \cdot m^{-3}$	<17	96	10	910	45	1200	1200	4700	240	53
$K_{f_{NM}}$	$ng \cdot m^{-3}$	40	15	3.8	2.6	1.2	1.7	4.0	45£	4.4	0.59
$Ca_{f_{NM}}$	$ng \cdot m^{-3}$	16	21	9.1	2.7	3.4	2.1	2.8	42£	7.3	2.2
Ti_f	$ng \cdot m^{-3}$	<2	<2.0	0.85	0.16	<0.063	0.22	0.17	0.37	<0.26	0.045
Fe_f	$ng \cdot m^{-3}$	28	26	6.4	1.1	0.54	1.4	1.3	3.3	4.0	0.41
Ni_f	$ng \cdot m^{-3}$	1	<0.36	0.08	<0.058	<0.027	<0.025	0.10	0.058	0.56	<0.032
Cu_f	$ng \cdot m^{-3}$	3	1.6		0.041	0.033	0.054	0.036	0.015	0.34	0.007
Zn_f	$ng \cdot m^{-3}$	9	2.6	0.18	0.14	0.11	0.23	0.084	0.071	0.93	0.036
Pb_f	$ng \cdot m^{-3}$	97	3.0	0.20	0.14	<0.097	0.30	0.056	0.14	1.56	<0.031

‡ Geometric mean of Aitken nuclei; • based only on 3 samples; § only sample 14; $ only sample 33; £ including marine component.
[1] Stevens et al., 1980; [2] Heintzenberg et al., 1981; [3] Flyger and Heidam, 1978 (total filters).

contaminated (nos. 4 and 23). Samples representative of seaspray, long-range transport and background aerosol have been averaged and are shown in separate columns.

Comparisons in Table 2 indicate that the *Ymer* grand average concentrations are about the same as summer Northern Greenland values except for chlorine which is much higher, due to the influence of the ocean. Arctic winter levels measured on Spitsbergen are one order of magnitude higher for parameters which are possible pollution indicators i.e. σ_{sp}, soot, non-marine S, Ni, Cu, Zn and Pb. Non-marine concentrations are obtained by subtracting the marine component calculated from bulk sea composition using Cl as tracer. It is interesting to note that the primary aerosol constituents soot, Fe, Ni, Cu, Zn and Pb have a larger winter to summer ratio than the secondary constituent S. Eastern United States rural summer concentration levels are one to two orders of magnitude higher than the *Ymer* levels. The largest differences are found for soot and lead while the lowest differences are found for components such as K, Ca, Ti and Fe which are often associated with soil erosion.

Size distributions of sulphur representing local contamination, Arctic summer background and long-range transport as measured in southern Sweden are plotted in Fig. 5. In some of the graphs the soot size distribution is also plotted.

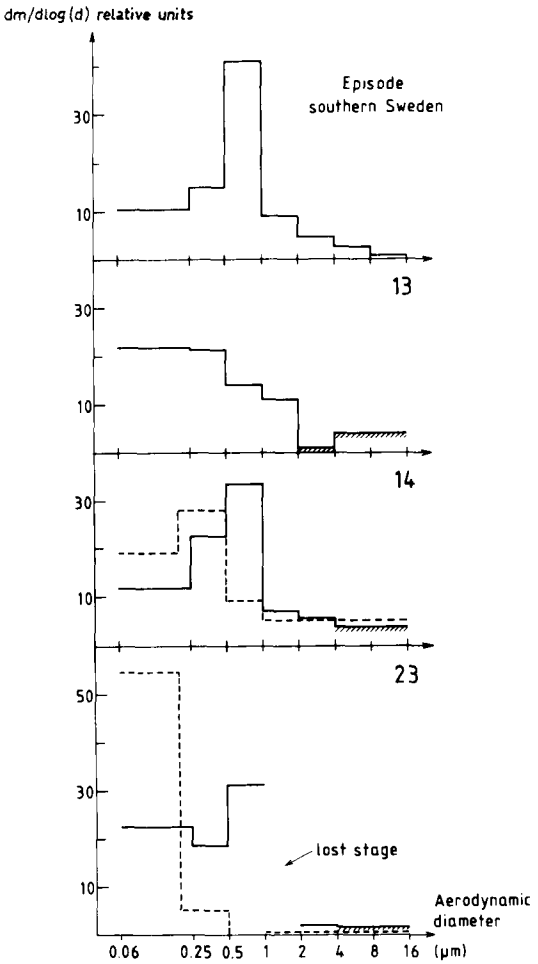

Fig. 5. Sulphur (——) and soot (– – –) size distributions taken during the *Ymer* expedition, compared with an average size distribution obtained from measurements in southern Sweden during episodic conditions, i.e. long-range transport from Western Europe. The size fractions of the cascade impactors include 2 open intervals: >8 μm and <0.25 μm. In order to present these intervals graphically, an upper size limit of 16 μm and a lower size limit of 0.06 μm were chosen. In the same way, 2 and 0.06 μm have been chosen as upper and lower size limits in the case of the soot distributions. ///// indicates detection limit. It should be pointed out that the column *area* reflects the relative mass load at each stage.

4. Discussion

The Arctic summer aerosol exhibited an unexpectedly large variability in size distribution and chemical composition. In order to better understand the cause of the variations, several sampling periods were selected for further evaluation. Thus we will discuss separately the characteristics of emissions from different sources, how they can be distinguished and how local contamination can be identified and accounted for.

4.1. *Indications of direct and indirect local contamination*

Atmospheric studies in the Arctic generally pose great demands on the sampling routines to ensure uncontaminated samples. When using a 22,000 horsepower icebreaker as a research platform, those demands must be set even higher. In order to be able to identify contributions from the icebreaker's diesel engines, total filter samples were taken from 1 of the 5 exhaust pipes. The com-

position is given in Table 3. The ratios between smoke and Arctic summer accumulation mode concentrations (see Table 3) are indicators of the dilution that has to take place to reduce each m^3 (of about 500 m^3 flue gases emitted/min) of the ship's emissions to Arctic summer concentrations. Since the exhaust concentrations also include coarse mode particles, the ratios should be regarded as upper limits, as if the smoke were only confined to accumulation mode particles.

The most sensitive indicators of the ship's emissions are those components having the highest ratios, i.e. soot and CN, while the metals are at least 2 orders of magnitude less sensitive to smoke contamination. The sulphate (as sulphur) ratio is of the same order of magnitude as those of the two metals detected. Since sulphate is gradually produced from its gaseous precursor, sulphur dioxide, the relative and absolute sulphate contribution in the smoke plume will change with the age of the plume. Therefore it is difficult to estimate the sensitivity of sulphate as a plume indicator. An instrument continuously measuring either CN or soot and operating with a short time constant would therefore be ideal to protect the air sampling on board. A recently developed CNC (see Table 1) fulfilled these criteria and was used to control the sampling system as described earlier.

Aerosol formation in emissions from high-temperature sources e.g. diesel combustion, results in very fine particles in the nucleation mode (particles wth a diameter <0.05 μm) growing through coagulation (Whitby, 1978) into the accumulation mode. High concentrations (measured by a CNC) of very fine particles resulting in a very small mass median diameter can therefore be used to identify freshly formed aerosols. The size distribution of soot (graphitic carbon) was measured in 4 stages (see Table 1) and can thus be used to identify sampling periods with possible contamination from the ships emissions. Very high soot concentration, of which 90% was found in particles with a diameter less than 0.2 μm, was experienced in sample 23. The typical relative soot fraction in the size range below 0.2 μm particle diameter is 30%, which is also the late winter average as measured at Spitsbergen (Heintzenberg, in press). On this basis, sample 23 was excluded as being contaminated. Of course this reasoning concerning soot (very-fine-particle concentration as

Table 3. *Measured total elemental and soot concentrations in emissions from one of the ship's 5 diesel engines; ratios to Arctic summer accumulation mode concentrations are also included*

Parameter	Diesel smoke concentrations[$]	Diesel smoke Arctic summer[1)]	Diesel smoke Arctic summer[2)]
CN (cm^{-3})	$1 \cdot 10^{9}$[§]	$5 \cdot 10^{6}$	$8 \cdot 10^{6}$
C_T	13,000		$3 \cdot 10^{6}$
S	400	15,000	4,000
Cl	<40	<4,000	<44
K	<30	<8,000	<12,000
Ca	<40	<4,500	<15,000
Ti	<10	<12,000	<65,000
Fe	<10	<1,500	<9,000
Ni	<0.5	<6,000	
Cu	0.5		13,000
Zn	1.0	5,500	7,000
Pb	<0.6	<3,000	<4,500

[$] in $\mu g \cdot m^{-3}$; [§] estimated.
[1)] Flyger and Heidam, 1978.
[2)] This work.

fresh aerosol indicator) also applies to other primary aerosol constituents e.g. Ni, Cu and Zn.

Indirect contamination i.e. aerosols emitted by the ship or the helicopters not directly entering the sampling system but allowed to age (thus transforming the particle size distribution towards larger particles) can probably not be identified using the above-mentioned criterium. However, using the ratios of metals to soot, one can infer that, if the soot values are reasonably low and therefore probably not contaminated, the metals are with an even higher probability uncontaminated. As mentioned earlier this reasoning may not be applicable to sulphate.

Garbage, mostly packaging materials, was burned on two occasions. Although placed on the leeward side, turbulence from the ship's body caused particulate emissions to contaminate the ship and the upper parts of the sampling line. When restarting sampling, some of this material became resuspended and contaminated especially the large particle collectors of the isokinetic samplers. As an indicator of such contamination the copper concentrations as measured by a total filter sampler are shown in Fig. 4, where the garbage fires are marked with G. In order to avoid further resuspension, the sampling pipe was taken apart and cleaned (C in Fig. 4). Unfortunately not all material was removed but rather loosened and resuspended during the subsequent sampling interval.

Aerosol blank samples i.e. air going through the main line but all sampling pumps switched off, were taken during 2 periods. Samples "exposed" this way are compared with non-exposed blanks in Table 4 to reveal whether aerosol particles enter the samplers during periods when the CN-level is exceeded and the sampling pumps are shut off. The results show additional material or blank content variation of the exposed blanks exceeding one standard deviation of the non-exposed blanks only for the elements S and Zn.

4.2. *Emissions from Arctic settlements*

Arctic settlements and human activities producing air pollution are insignificant compared with midlatitudinal sources. However, the remoteness of the Arctic in combination with the summer decoupling can make the anthropogenic emissions within the Arctic region important for the summer concentration levels found. Jaenicke and Schütz (1982) propose the settlements and related activities to be the main source of condensation nuclei (CN) during the Arctic summer.

A close study of those sampling periods having comparably high and relatively invariable CN concentrations reveals that they are taken mostly during periods when ground level trajectories (Jaenicke and Schütz, 1982) indicate that Arctic settlements can have contributed to the aerosol concentration. Examples of such samples are 6, 8, 24, 25, 27 and 28 while samples 14 and 22, although having relatively high CN concentrations can be related to long-range transport and sea-spray, respectively, as will be discussed later. A

Table 4. *Comparison of "exposed" (fan sucking air through main line, samples in position but sampler pumps switched off) and non-exposed blanks for elements detected*

Parameter	"Exposed" samples 26	"Exposed" samples 36	Non-exposed blank Average ± one sigma
TSP	1.70095[§]	1.68031[§]	1.7011[§] (26) 1.67942[§] (36)
C_T	0.036*	0.037*	0.038*
S	45	52	44 ± 4.8
Cl	29	12	25 ± 5.8
Ca	7.4	11	10 ± 2.1
Fe	9.9	8.7	9.4 ± 1.0
Ni	<0.42	0.54	0.47 + 0.080
Cu	<0.42	0.55	0.56 ± 0.11
Zn	0.36	0.43	0.37 ± 0.023

[§] Filter weight in g; * optical density measured in the soot photometer.

comparison with the other parameters in Fig. 4 shows that high CN values do not in general correspond to high values of light scattering, soot or fine particle elemental concentrations. This implies that Arctic sources do not contribute a large amount to the light scattering, soot or elemental concentrations i.e. these emissions do not drastically alter the accumulation mode aerosol composition.

4.3. *Seaspray*

Seaspray is generated when air bubbles mixed in the water ascend and traverse the surface, thus producing droplets of different sizes (see e.g. Blanchard and Woodcock, 1980). Using a ship as a sampling platform will add sea spray droplets produced from splashing at the bow, even when breaking ice. 2 sampling periods were found to be mainly influenced by sea spray, one representing mainly the first production mechanism while the second is a mixture of both mechanisms. Sample 22 was taken just north of Kong Karls land in open water at one position and with an average wind speed of 12 $m \cdot s^{-1}$. Sample 33 was taken mainly in heavy pack ice while the ship was moving north west of Franz Joseph land. The average wind speed was 6 $m \cdot s^{-1}$.

Characteristics of sea spray as found in those two samples (see Table 2) are: high TSP, σ_{sp}, Cl, K and Ca and normal CN, C_T and most of the metal concentrations. As shown both by σ_{sp} and the accumulation mode elemental concentrations, sea salt contributes considerably to the accumulation mode aerosol mass. The TSP concentrations are lower than predicted by Lovett (1978) for 15 m above the sea surface, but can mostly be explained by sea salt using Cl as a tracer and adding proportionally Na, Mg, S, K and Ca. In Table 5, ratios to the sea salt indicator chlorine are calculated for samples 22 and 33; S, K, Ca and Br to Cl ratios are on the same level as for bulk sea water compositon. However, in sample 33 Br shows some deficit while there is some evidence for an additional contribution of sulphur in both samples. The heavy metals all have ratios several orders of magnitude too high and therefore must have their main contributions from other sources.

4.4 *Possible influence of long-range transport*

According to earlier discussions, the Arctic region is more or less decoupled and should therefore experience few occurrences of long-range transport from midlatitudinal source areas during the summer. Trajectories calculated by the Norwegian Meteorological Institute to Northern Norway,

Table 5. *Sea salt characteristics and composition relative to chlorine compared with bulk sea water composition for the accumulation + coarse fractions of the 2-stage filter sampler*

Parameter	Sample 22	Sample 33	Bulk sea
Sea salts[$] ($\mu g \cdot m^{-3}$)	30	10	
TSP ($\mu g \cdot m^{-3}$)	19	3.9	
CN (cm^{-3})	460	89	
σ_{sp} ($10^{-6} \cdot m^{-1}$)	[&]	4.8	
C_T ($ng \cdot m^{-3}$)	5.3	5.6	
Cl ($ng \cdot m^{-3}$)	7,800	1,600	
Cl/TSP	0.41	0.41	0.55
S/Cl	0.075	0.063	0.047
K/Cl	0.028	0.017	0.021
Ca/Cl	0.027	0.021	0.021
Fe/Cl	0.0021	0.0030	$7 \cdot 10^{-7}$
Zn/Cl	0.00042	0.00023	$7 \cdot 10^{-7}$
Br/Cl	0.0028	0.0015	0.0035
Pb/Cl[§]	$8 \cdot 10^{-5}$	$1 \cdot 10^{-4}$	$2 \cdot 10^{-9}$

[$] Lovett (1978); [&] nephelometer not working; [§] accumulation mode.

Table 6. *Average (October 1978–September 1981) relative part of the time when trajectories on the 850 mbar level originating in central Europe reached the receptor-stations*

Location	July	August	September	
Jergul, Northern Norway	14	9	8	(%)
Bear Island	4	9	4	
Ny-Ålesund, Spitsbergen	1	4	2	

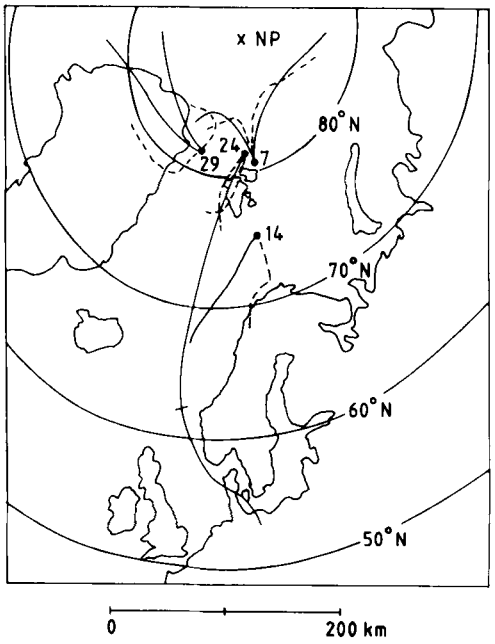

Fig. 6. Isobaric 48 h trajectories calculated at the surface (– – –) and at the 850 mbar level (——) by Schütz for samples 7, 14, 24 and 29. In cases where a significant change in the trajectory direction was observed, trajectories representing the start and end of the sampling period have been plotted. The sample 24, 850 mbar trajectory (72 h) was calculated by the Norwegian Meteorological Institute and closely resembles that calculated by Schütz (for the first 48 h).

Bear Island and Spitsbergen show that these areas are only exposed to air parcels originating in central Europe during a minor part of the time in the summer (see Table 6). Average concentrations (see Table 2) of soot and the heavy metals (except Ni) were highest in August. This might be related to the higher frequency of central European origin trajectories experienced in August at both Spitsbergen and Bear Island (Table 6). During the *Ymer-80* expedition only 2 or possibly 3 occurrences of long range transport were observed. It was not possible to clearly identify these events during the expedition. The first event took place at the very end of the expedition's first leg just after having left the working area as shown in Fig. 1. The duration of this episode was only a few hours during the night of the fourth and fifth of August. Corresponding surface and 850 mbar back trajectories calculated by Schütz (Jaenicke and Schütz, 1982) are shown in Fig. 6. Sample 14 surface trajectories indicate possible contributions from Northern Norway while the 850 mbar trajectory indicates possible contributions from further south. The second episode was experienced north east and north of Spitsbergen during sampling periods 23 and 24 on the second leg. As mentioned earlier, 23 is probably contaminated by emissions from the ship while 24 shows no indications of such contamination. However, as seen in Fig. 6 the air masses reaching *Ymer* during sample period 24 have passed over Spitsbergen. According to the earlier discussion, Arctic sources cannot be responsible for the comparably high levels of C_T, non-marine S, Ni, Cu, Zn and Pb measured during this period. Indications of long-range transport from air mass history, with a possible origin in European Russia were found for samples 31 and 32. High levels of σ_{sp}, C_T and non-marine S supported this hypothesis. However the sea spray concentration was also very high, complicating the identification of an aerosol long-range transport component.

Characteristics of "long-range transport" as measured in samples 14 and 24 (see Table 2) are: high levels of C_T, non-marine S and metal concentrations with elevated levels of CN, TSP and σ_{sp}. In addition increased levels of radon and radon-daughters reported by C. Samuelsson, Department of Radiophysics, University of Lund (unpublished data), indicate a continental non-permafrost origin of the air masses. The relative increase in concentration levels during a long-range transport episode compared with the Arctic "background levels" (discussed below) and the grand average are listed in Table 7. TSP, σ_{sp}, C_T, non-marine S, Ni, Cu, Zn and Pb have the highest ratios and are thus the most sensitive pollution

Table 7. *Relative increase in long-range transport (LRT) concentrations compared with Arctic "background levels" (ABL) and the grand average (GA), respectively; accumulation mode concentrations are indexed with f, coarse mode with c and the non-marine component with NM*

Ratio	CN	TSP	σ_{sp}	C_T	$S_{f_{NM}}$	Cl_{f+c}	K_{NM}	Ca_{NM}	Ti	Fe	Ni	Cu	Zn	Pb
$\frac{LRT}{ABL}$	4.9	18	13	41	23	4.5	7.5	3.3	<5.8	9.8	>18	49	26	>50
$\frac{LRT}{GA}$	3.0	1.6	6.0	12	3.7	0.26	1.7	2.7	<1.6	3.6	>9.7	8.3	6.6	11

indicators of long-range transport influence. Sea spray can contribute considerably to TSP and σ_{sp} as seen previously. Therefore they are not useful as such indicators.

A comparison of 4 sulphur and 2 soot size distributions is presented in Fig. 5. Sample 14 representing long-range transport has a definite peak in the 0.25–1 μm particle diameter range for both sulphur and soot. A sulphur peak in the same interval can also be discerned in sample 23 which in addition also has finer sulphur-containing particles, probably originating from the ship's emissions, as proposed earlier. This is supported by the soot size distribution having 90% of the concentration of particles finer than 0.2 μm diameter. In fact the sulphur size distribution can be interpreted as a combination of 2 size distributions with mass median diameters in the intervals <0.25 μm and 0.5–2 μm, respectively. Sample 13 taken immediately before 14 has a quite different, flatter size distribution. It is also interesting to note the resemblance of the long-range transport sulphur size distribution obtained in southern Sweden to that obtained in the Arctic (no. 14).

4.5. *Arctic background levels*

Background levels are often referred to as baseline concentrations when discussing the degree of pollution at a particular measuring site. Here we propose an operational definition that the samples having the lowest concentration levels and least risk of contamination represent the Arctic "background levels". Samples 7 and 29 fulfil these requirements. The main part of sample 7 was taken in air masses circulating around a weak high pressure center over the North Pole and represents an extremely aged air mass. In Fig. 6, trajectories representing the onset and end of the sampling period show that the trajectories gradually turned from north east to north west. The ship was situated north east of Spitsbergen. The second "background" sample (no. 29) was taken in air masses traversing North Eastern Greenland (see Fig. 6) during sampling off the North Eastern Greenland coast. The average background levels taken as the mean of samples 7 and 29 are compared with levels experienced during long-range transport and the grand average in Table 7. The background levels of potential air pollutants such as C_T, S, Ni, Cu, Zn and Pb are between 1 and 2 orders of mangitude lower than during episodic conditions. The difference compared with the grand average is generally one order of magnitude.

The variations of CN concentration and σ_{sp} during sampling period 29 can be studied in detail in Fig. 7. Rather high CN values were experienced at the onset of sampling when some influence from Station Nord (a Danish military base situated in North Eastern Greenland) caused the sampling to be shut off for a few hours on the morning of August 31. As pointed out in the diagram, passing banks of fog (C in Fig. 7) causes the CN concentration to drop one order of magnitude or more, reaching extremely low values of only a few particles per cm^3. The variations of σ_{sp} are within one order of magnitude, roughly between 10^{-7} and $10^{-6} \cdot m^{-1}$, which is much less than the CN concentration variations. The correlation between CN and σ_{sp} is not very high as can be seen in Fig. 7. It should also be noted that the influence from Station Nord cannot be traced in σ_{sp}, which confirms the previous reasoning and anticipations of the minor risk of accumulation mode contamination from Arctic settlements.

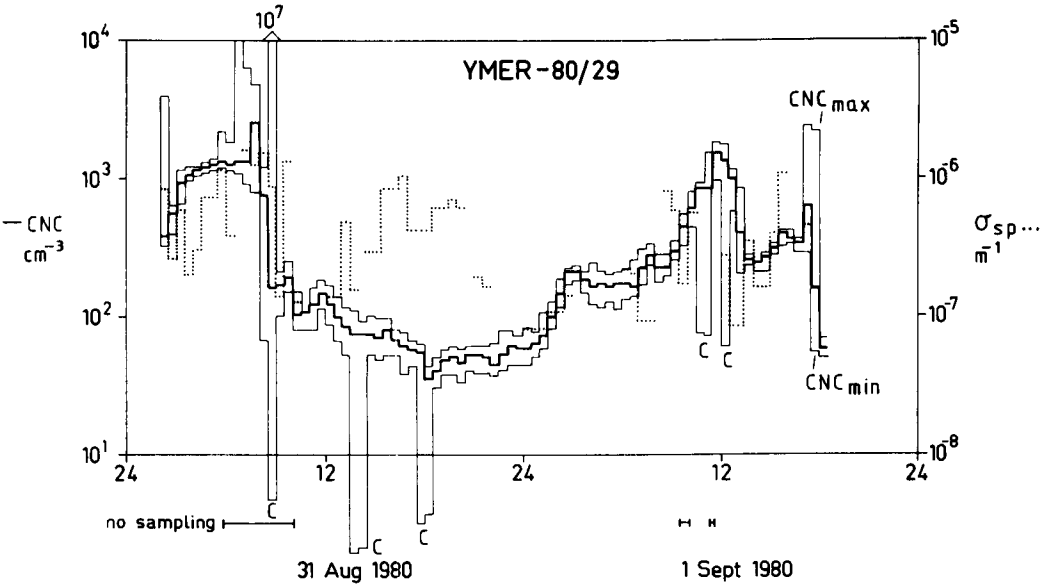

Fig. 7. Variation of CN-concentration and σ_{sp} of sample 29. The area between maximum and minimum CN-values for each 30 min sampling period is marked as a shaded area about the median (strong line). The time resolution of the nephelometer is about 30 min. The occurrence of fog banks or precipitation is marked with a C.

5. Summary and conclusions

As a consequence of a strict adoption of Rahn's coupling-decoupling hypothesis, low and constant levels of especially the anthropogenic aerosol components should be expected in the Arctic during the summer season. The Arctic summer ground level aerosol composition exhibited variations for most of the measured physical and chemical parameters of 1 to 2 orders of magnitude. The variation time constant appeared to be of shorter duration than the measurement periods which were typically 1 to 2 days. Much of the variations can be explained by influence from aerosol long-range transport, seaspray and settlement emissions in combination with the varying strength of scavenging mechanisms. The characteristics of different source types are summarized in Table 8.

From the experience gained in this study we

Table 8. *Rough characteristics of different source types influencing the Arctic aerosol composition*

Parameter	Ship's emissions	Settlement emissions	Long range transport	Sea spray
CN	H	H	I	I
TSP		L	I	H
σ_{sp}		L	H	H
C_1	H	L	H	I
S_{NM}	H	L	H	H*
Cl	L	L	L	H
Heavy metals	H§	L	H	I or L

* Including marine component; § Cu and Zn.
H = high, I = intermediate and L = low level.

conclude that the mysterious background aerosol often referenced as a hypothetical lower limit content aerosol, does not exist. The lowest levels measurable i.e. the "background aerosol" are just a function of source and sink mechanisms in combination with obtainable measurement time resolution.

6. Acknowledgements

We acknowledge the support from the *Ymer-80* expedition represented by its scientific leader Professor Valter Schytt. The commanding officers made a great effort to maintain good air sampling conditions which is gratefully acknowledged. We are very much indebted to Leif Bäcklin who set up the laboratory containers used on the expedition and to Erik Karlsson and Lars-Berne Nilsson who modified and built the samplers used in conjunction with the PIXE-analysis. We thank Dr. Lothar Schütz and the Norwegian Meteorological Institute for providing the trajectories. This project was supported by the Swedish Natural Science Research Council.

REFERENCES

Blanchard, D. C. and Woodcock, A. H. 1980. The production, concentration and vertical distribution of the sea-salt aerosol. Aerosols: anthropogenic and natural, sources and transport (eds. T. J. Kneip and P. J. Lioy). *Annals of the New York Academy of Sciences 338*, 330–347.

Bricard, J., Delattre, P., Madelaine, G. and Pourprix, M. 1976. Detection of ultrafine particles by means of a continuous flux condensation nuclei counter. In *Fine particles* (ed. B. Y. H. Liu). Academic Press, New York, 565–580.

Cahill, T. A., Eldred, R. A., Barone, J. and Ashbaugh, L. 1979. Ambient aerosol sampling with stacked filter units. Prepared for Federal Highway Administration, Offices of Research & Development, Washington, D.C. 20590, U.S.A. Report no. FHWA RD-78-178.

Central Intelligence Agency 1978. *Polar Regions Atlas.* U.S. Govt. Printing Office, Stock no 041-015-0094-2, Washington, D.C., U.S.A.

Charlson, R. J., Porch, W. M., Waggoner, A. P. and Ahlquist, N. C. 1974. Background aerosol light scattering characteristics: nephelometric observations of Mauna Loa Observatory compared with results at other remote locations. *Tellus 26*, 346–360.

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

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

Heintzenberg, J., Hansson, H.-C. and Lannefors, H. O. 1981. The chemical composition of arctic haze at Ny-Ålesund, Spitsbergen. *Tellus 33*, 162–171.

Heintzenberg, J. 1982. Measurement of light absorption and elemental carbon in atmospheric aerosol samples from remote locations. In *'Particulate Carbon Atmospheric Life Cycle'* (eds. G. T. Wolff and R. L. Klimisch). Plenum Publication Co., New York, N.Y. 100 13, U.S.A.

Heintzenberg, J. 1982. Size-segregated particulate elemental carbon and aerosol light absorption at remote arctic locations. *Atmos. Environ.*, in press.

Jaenicke, R. and Schütz, L. 1982. Arctic aerosols in surface air. *Idöjaras 86*, 235–241.

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.

Lovett, R. F. 1978. Quantitative measurement of airborne sea-salt in the North Atlantic. *Tellus 30*, 358–364.

Mitchell, R. I. and Pilcher, J. H. 1959. Improved cascade impactor for measuring aerosol particle sizes in air pollutants, commercial aerosols and cigarette smoke. *Ind. Eng. Chem. 15*, 1039–1042.

Rahn, K. A. and Shaw, G. E. 1978. Briefing on Arctic haze and the Arctic aerosol. Presented to the Bureau of Oceans and International Environmental and Scientific Affairs. Department of State, Washington, D.C. 20550, U.S.A., 20 November 1978.

Rahn, K. A. and McCaffrey, R. J. 1980. On the origin and transport of the winter Arctic aerosol. Aerosols: anthropogenic and natural, sources and transport (eds. T. J. Kneip and P. J. Lioy). *Annals of the New York Academy of Sciences 338*, 486–503.

Stevens, R. K., Dzubay, T. G., Shaw, R. W., McClenny, J. W. A., Lewis, C. W. and Wilson, W. E. 1980. Characterization of the aerosol in the Great Smokey Mountains. *Environ. Sci. Tech. 14*, 1491–1498.

Whitby, K. T. 1978. The physical characteristics of sulphur aerosols. *Atmos. Environ. 12*, 135–159.