

## ARCTIC AIR POLLUTION: AN OVERVIEW OF CURRENT KNOWLEDGE

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**Abstract**—From December to April, the Arctic air mass is polluted by man-made mid-latitude emissions from fossil fuel combustion, smelting and industrial processes. In the rest of the year, pollution levels are much lower. This is the outcome of less efficient pollutant removal processes and better south (S) to north (N) transport during winter. In winter, the Arctic air mass covers much of Eurasia and N. America. Meteorological flow fields and the distribution of anthropogenic SO<sub>2</sub> emissions in the northern hemisphere favor northern Eurasia as the main source of visibility reducing haze. Observations of SO<sub>4</sub><sup>2-</sup> concentrations in the atmosphere throughout the Arctic yield, depending on location and year, a January–April mean of 1.5–3.9 μg m<sup>-3</sup> in the Norwegian Arctic to 1.2–2.2 μg m<sup>-3</sup> in the N. American Arctic. An estimate of the mean vertical profile of fine particle aerosol mass during March and April shows that, on average, pollution is concentrated in the lower 5 km of the atmosphere. Not only are anthropogenic particles present in the Arctic atmosphere but also gases such as SO<sub>2</sub>, perfluorocarbons and pesticides. The acidic nature and seasonal variation of Arctic pollution is reflected in precipitation, the snowpack and glacier snow in the Arctic. A pH of 4.9–5.2 in winter and ~5.6 in summer is expected in the absence of calcareous wind-blown soil. Glacial records indicate that Arctic air pollution has undergone a marked increase since the mid 1950s paralleling a marked increase in SO<sub>2</sub> and NO<sub>x</sub> emissions in Europe. Effects of Arctic pollution include a reduction in visibility and perturbation of the solar radiation budget in April–June. Potential effects are the acidification and toxification of sensitive ecosystems.

### INTRODUCTION

It has been approximately 30 y since the first documented report of Arctic air pollution and the coining of the term 'Arctic haze' by Mitchell (1956). Based on the qualitative observations of weather officers aboard the routine 'Ptarmigan' weather reconnaissance flights in the Alaskan Arctic, he made several observations: (i) haze is comprised of particles no larger than 2 μm diameter that are not ice crystals; (ii) it occurs in patches some 800–1300 km across; (iii) it has been observed at all levels in the atmosphere below ~9 km; and (iv) curiously, it is seldom found over Greenland.

These observations would lie dormant, unused by the atmospheric physics and chemistry community, for almost 20 y before they were linked with wind-blown dust and man-made air pollution. Serendipity eventually prevailed. Unexpectedly high values of total atmospheric turbidity measured in the Alaskan Arctic during spring of 1971 and 1972 (Shaw and Wendler, 1972) prompted a renewed interest in Arctic haze. No one knew much about its nature and origin. A subsequent study of the vertical profile of turbidity (Holmgren *et al.*, 1974) found layers of anomalously high turbidity at altitudes of a few kilometers. About 40% of the total turbidity was above 4 km. This, in turn, was the motivation for an aircraft study of the chemical composition of haze particles on 15 flights during April and May 1976 (Rahn *et al.*, 1977). It was found that although particulate matter in air between

distinct haze layers was pollution-derived (as indicated by enrichment in oil-derived vanadium), the haze layers themselves were of crustal composition originating presumably from Asian deserts between 40 and 50°N. This study was unjustifiably interpreted by many to mean that Arctic haze consisted mainly of wind-blown dust. It was not until the first routine ground-level observations of the chemical composition of suspended particulate matter in the winter of 1976–1977 (Rahn and McCaffrey, 1979a, 1980) that another picture emerged. Arctic haze particles in the lower troposphere of the Alaskan Arctic were found to undergo a very strong seasonal variation. They were 20–40 times more abundant in winter than in summer. Furthermore, during winter they were mostly of man-made origin while in summer, what little there was consisted of wind-blown dust and sea salt.

The investigations of Rahn and McCaffrey (1979a, 1980) and the first symposium on Arctic aerosols held in April 1977 in Norway initiated a revolution in Arctic air pollution research. This included the establishment of an international Arctic aerosol sampling network (Rahn, 1981). The state of knowledge in 1981 was summarized by Rahn and Heidam (1981). In particular, it was concluded that: (i) the entire Arctic is exposed to pollution aerosol of distant origin during winter; (ii) the two most important pathways of pollution transport from mid-latitudes are from Eurasia; and (iii) the average composition of pollution aerosol during winter/spring is 2 μg m<sup>-3</sup> SO<sub>4</sub><sup>2-</sup>,

$1 \mu\text{g m}^{-3}$  organic compounds,  $0.3\text{--}0.5 \mu\text{g m}^{-3}$  black carbon, a few tenths of a  $\mu\text{g m}^{-3}$  of other substances and a few  $\mu\text{g m}^{-3}$  of water.

Since 1981 new insight has been gained into pollutant pathways and vertical distributions in the Arctic. In this overview, the current state of our knowledge of Arctic air pollution and the literature from which this knowledge is drawn is summarized. Previous reviews of Arctic haze have been published which the reader is encouraged to consult for additional background information (Rahn and McCaffrey, 1979a; Rahn, 1982a, b). Furthermore, three important compendia of Arctic research results are found in the Arctic Symposia (1981, 1985) and in AGASP (1984).

### THE POLLUTION METEOROLOGY OF THE ARCTIC

The Arctic air mass is a unique meteorological feature of the troposphere characterized by: (i) sub-zero temperatures much of the year; (ii) little precipitation (most of which falls during the warmer half of the year); (iii) stable stratification that prevents strong vertical mixing; and (iv) low levels of solar radiation especially during winter. During winter, it occupies, in addition to the polar region, a large fraction of the snow-covered continental land mass of N. America and Eurasia. This is illustrated in Fig. 1 where the

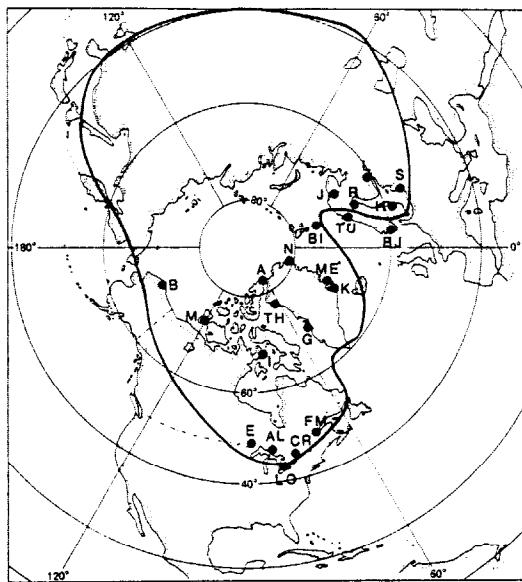


Fig. 1. The mean position of the Arctic front in January (solid line) from Barrie and Hoff (1984) and the location of air monitoring stations providing observations used in Figs 8 and 9.

A-Alert	AL-Algoma	B-Barrow
BI-Bear Island	BJ-Birkenes	CR-Chalk River
E-ELA-Kenora	FM-Forêt Montmorency	G-Godhavn
H-Hoburg	I-Igloolik	J-Jergul
K-Kap Tobin	L-Lesogorski	LO-Long Point
M-Mould Bay	ME-Mesteruig	N-Nord
R-Ricklea	S-Suwalki	TH-Thule
TU-Tustervaten		

mean position of the Arctic front at the surface is depicted for January (Barrie and Hoff, 1984). In three dimensions, the air mass at this time of year consists of a dome 7–8 km deep over the pole with shallow tongues of air 0–5 km deep spilling southward over the land mass. When Arctic air intrudes southward over the northern ocean, strong modification over relatively warm water rapidly alters its meteorological characteristics. On the other hand, southward intrusions over cold snow-covered land are not altered so rapidly.

The Arctic air mass is very different in summer than in winter. In summer, it is confined to the northern polar region and has more cloud than in winter. More cloud cover in summer is associated with a greater abundance of stratus clouds in the lower troposphere. This is well illustrated by the annual variation in occurrence of lower (0–2 km), middle (2–4 km) and upper (4–7 km) level clouds as well as clear skies in the western Eurasian Arctic (Fig. 2) reported by Huschke (1969). While low cloud frequency undergoes a marked seasonal variation ranging from ~75% between May and September to ~35% between December and March, middle and high cloud frequencies vary less. The frequency of clear skies is very low in summer (5–8%) and relatively high in winter (30–40%). These results are typical of the Arctic region. The spatial distribution of mean monthly low cloud cover in January and July is shown in Fig. 3. In July, it ranges from a maximum of 75% over the Arctic ocean to 40–50% in the N American and Greenland sectors of the Arctic. In January, the frequency of low cloud is 20–35%. It has a marked maximum over the Norwegian and Barents seas off northern Scandinavia and a minimum in the high Arctic of the eastern Soviet Union.

Since low level stratus in summer is often formed by warm air advection over the cold ice pack (Tsay and Jayaweera, 1984), it is frequently associated with drizzle. Hence, it is important as a means of removal of

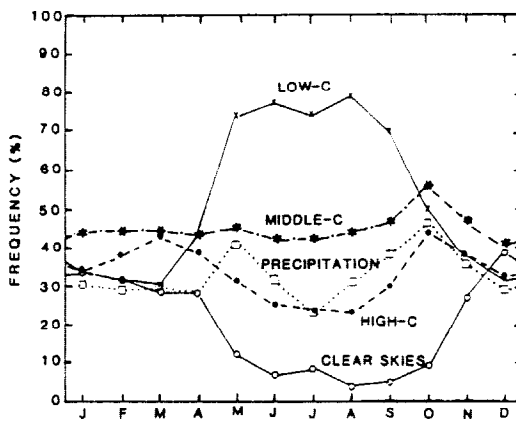


Fig. 2. The temporal variation of mean monthly frequencies of low, middle and high cloud, precipitation and clear skies in the Western Eurasian Arctic (from Huschke, 1969).

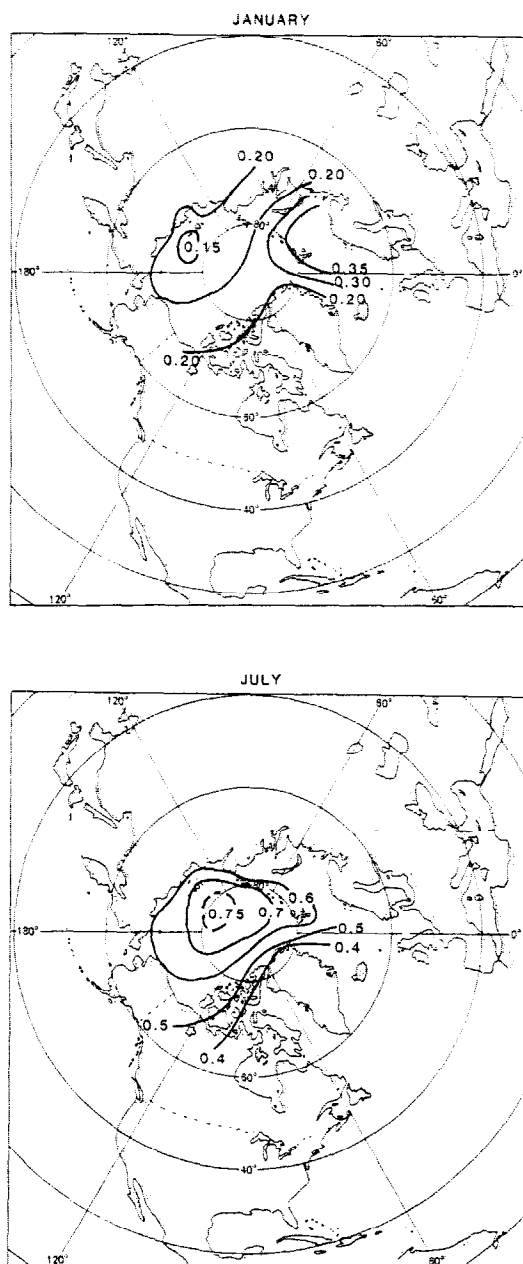


Fig. 3. The spatial distribution of the estimated mean fraction of sky covered by low cloud in January and July in the Arctic (from Huschke, 1969). This parameter is also approximately equal to the frequency of time that low cloud is observed.

air pollution. Indeed, it is observed that the annual variation of low level cloud cover is in anti-phase to the annual variation of anthropogenic  $\text{SO}_4^{2-}$  concentration in the lower Arctic troposphere (Barrie *et al.*, 1981). An investigation of Arctic cloudiness since 1920 by Raatz (1981) has revealed no significant trends that might be ascribed to increasing levels of air pollution. Of course, such studies are limited to a certain extent by: the paucity of meteorological observations in the N, the unknown sensitivity of cloud cover to pollution influences and the large natural variability of cloud

cover caused by variations in global circulation patterns.

#### POLLUTION SOURCES AND TRANSPORT PATHWAYS

An overview of Arctic haze would be incomplete without considering the location of pollutant sources relative to the Arctic air mass and to major features of the atmospheric circulation pattern of the northern hemisphere. Nature and man have conspired to make northern Eurasian sources far more available to the Arctic than those in N. America. Since  $\text{SO}_4^{2-}$ , originating from  $\text{SO}_2$  produced in the burning of fossil fuels and smelting of sulphide ores, is a major anthropogenic constituent of haze particles ( $\sim 30\%$ ; Barrie *et al.*, 1981) a source inventory of annual  $\text{SO}_2$  emissions was constructed (Fig. 4). It was done for those areas that are in Arctic air at some time during the winter. North American emissions were obtained from estimates put together by the U.S./Canada Working Group on Acid Rain (see, for instance, Barrie and Hales, 1984). Emissions west (W) of longitude  $60^\circ\text{E}$  were obtained from a recent report (EMEP/CCC, 1984) of the European monitoring program. Note that Soviet emissions east (E) of longitude  $60^\circ\text{E}$  are very uncertain. They were obtained by apportioning the better known emissions for the eastern Soviet Union roughly according to population and industrial activity. Izrael (1984) reports that total  $\text{SO}_2$  emissions in the Soviet Union in 1984 were  $23 \times 10^6$  tonnes (23 Mt). It is also known that annual Soviet emissions W of  $65^\circ\text{E}$  are approximately 16 Mt (EMEP/CCC, 1984). This leaves 7 Mt for the areas of Norilsk, eastern Siberia and Kazakhskaya in the central S. Recent

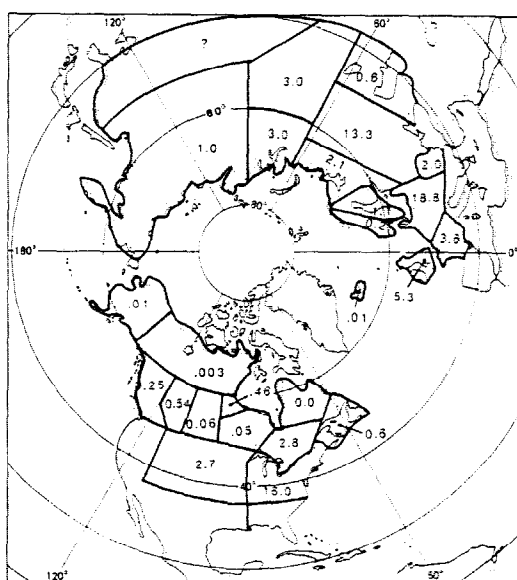


Fig. 4. Annual emissions of  $\text{SO}_2$  (Mt) in regions of the northern hemisphere that influence the Arctic. Representative of the early 1980s.

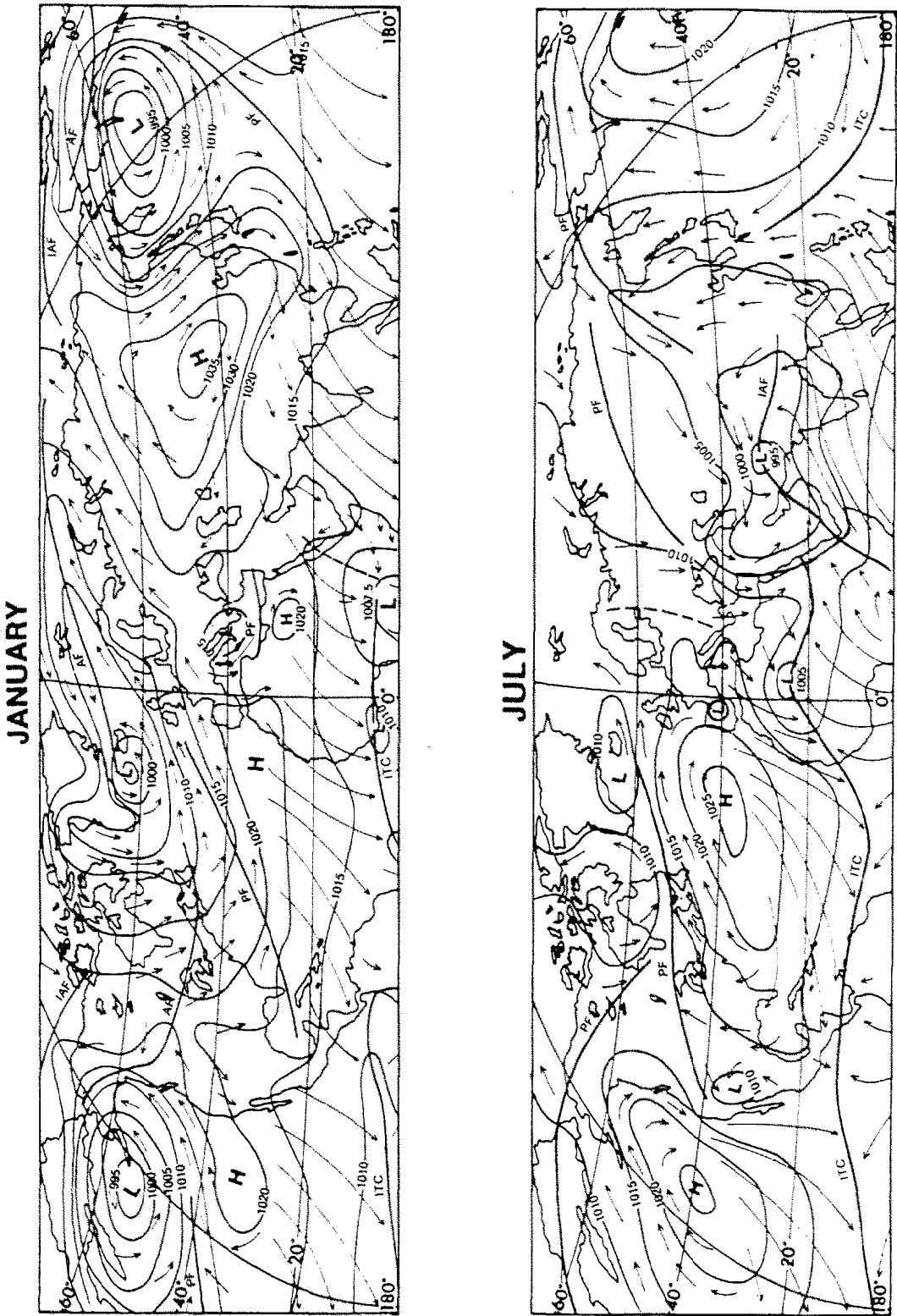


Fig. 5. Average values of surface pressure (thin lines; unit mb), winds (long arrows, steady winds; short arrows, unsteady winds), frontal zones and convergence zones in the northern hemisphere during January and July (from Lijquist, 1970).

unpublished information indicates that the estimate of 16 Mt W of 65°E may be about 20% high. This would mean that an even larger fraction of emissions occurs east of the 65°E. At this time, the author is unaware of published emission estimates for Mongolia or northern China. However, these sources are likely of little importance for the Arctic since they are far S and separated from the N by mountains.

Eurasian SO<sub>2</sub> sources that are available to the Arctic in winter are more than double those in N. America ( $54 \times 10^6$  vs  $24 \times 10^6$  t y<sup>-1</sup>). The difference is even greater in the region N of 60° latitude (6.3 vs 0.013 Mt y<sup>-1</sup>). This reflects the relative populations of northern Eurasia and northern N. America. In the absence of volcanic emissions, natural sources of SO<sub>2</sub> in the Arctic region during winter are likely to be small. Biogenic sources will be low due to ice and snow cover. The smoking hills near the Mackenzie delta in northern Canada are natural lignite deposits that have spontaneously combusted (Hutchinson *et al.*, 1978). Total annual SO<sub>2</sub> emissions are estimated to be approximately 50 t y<sup>-1</sup> (Farg, 1977). Occasionally, a volcano erupts in or near the Arctic air mass and can provide substantial amounts of natural SO<sub>2</sub> to the atmosphere. Evidence of the eruption of Mt. Katmai in the Aleutian islands in 1912 has been seen in the acidity of glacial ice cores (Barrie *et al.*, 1985). Volcanic influences later in this century are not evident.

How does the anthropogenic emission of SO<sub>2</sub> in the northern hemisphere fit into the wind patterns? To help answer this question, consider the mean flow field in the lower troposphere in January and July that is shown in Fig. 5. The mean surface pressure features and the mean position of the Arctic and polar fronts are also shown. In both summer and winter, the mean flow in N. America is SE. In Eurasia, it is SE in summer but strongly NE in winter.

In winter, when the Arctic is most polluted, the predominant pathway of windborne pollution from the high emission areas of eastern N. America to the

pole is E over the stormy Atlantic. In contrast, Eurasian pollution travels NE over snow covered land-ice areas. There is a marked difference in the precipitation encountered along these respective pathways. This was most vividly depicted by Rahn (1982a) in Fig. 6. Note that the cumulative mean monthly precipitation encountered by N. American and SE Asian pollution on its most probable route to the Arctic is at least four times greater than that encountered by Eurasian sources.

The strong transport into the Arctic from Eurasia during winter is caused by the presence of the climatologically persistent Siberian high pressure region. Like energy transfer (Oort, 1974), mass transfer takes place between the Arctic and southerly latitudes by the action of transient and stationary large-scale eddies the size of high and low pressure regions. In a detailed analysis of synoptic situations leading to strong transport out of Eurasia and N. America into the Arctic, Raatz and Shaw (1984) were able to confirm that the link between the Arctic and N. America is weak while the Eurasian connection was strong. 'Surges' of air N out of Eurasia during winter were associated with synoptic situations that typically involved the tracking of a low pressure cell from Iceland into the Murmansk area or across Europe until it encounters the semi-persistent Siberian high. At this point, a northward flow of air associated with a strong E-W pressure gradient between the low and high pressure zones is maintained for several days. This results in a surge of polluted European air into the Arctic. Upper air circulation patterns at 500 mb often support these surges. Raatz and Shaw (1984) classified surges into two groups according to the area from which air was drawn either from eastern Europe (mainly Soviet Union) or central and western Europe (including Scandinavia). The frequency of occurrence of each type of surge for the winter months when pollution is at its peak in the Arctic is shown in Table 1. As winter progresses, the area from which air is drawn in Europe

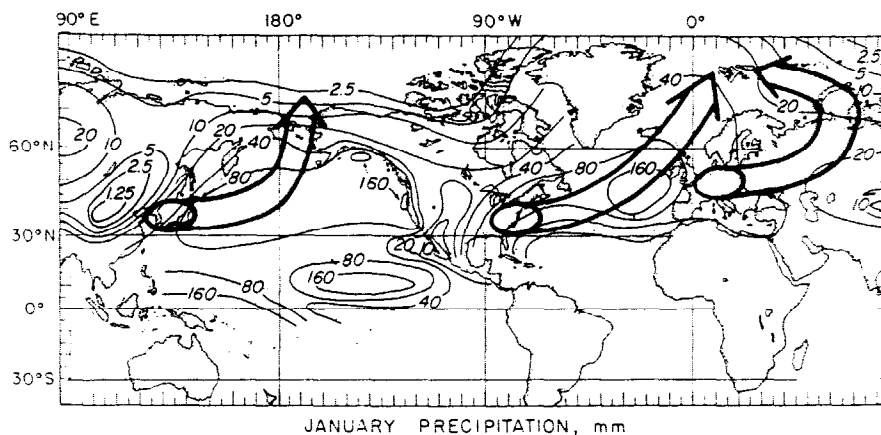


Fig. 6. Spatial distribution of mean monthly precipitation in January (mm of water). Arrows indicate the predominant pathway of air out of major pollutant source regions to the Arctic (from Rahn, 1982a).

Table 1. The frequency of occurrence of surges of air from eastern Europe (including the Soviet Union W of the Urals) and western Europe during winter (extracted from Raatz and Shaw, 1984)

Month	Number of surges	
	E	W
October	2	1
November	8	0
December	5	4
January	5	3
February	4	6
March	0	7

shifts from E to W. Confirmation of this qualitative analysis of pollution sources is needed before one can be sure that this is indeed the case. Chemical transport models and elemental tracer studies will prove useful in doing this.

During the AGASP flight period of March 1983, a surge of air out of Eurasia occurred around 5 March and arrived in Barrow, Alaska on 11 March (Raatz, 1984; Harris, 1984). It was supported by upper air flow patterns at 500 mb. Detailed air chemistry measurements were made during this period (AGASP, 1984).

An estimate of trace element emissions from anthropogenic sources in Europe was made by Pacyna (1984). Based on his calculations, a comparison of emissions from different parts of the region which are the most likely source of aerosols in the Arctic was made (Table 2). The emissions from western, central and eastern Europe are for the most part, within a factor of two of one another. Central European emissions tend to be somewhat lower than those from western Europe and the Soviet Union. This information may prove useful in confirming the exact source of Eurasian aerosols.

#### OBSERVATIONS OF ARCTIC AIR POLLUTION

##### *Occurrence of particulate pollution*

The temporal and spatial distribution of anthropogenic aerosols in the Arctic atmosphere has been under routine investigation at ground level for a number of years. The seasonal variation of anthropogenic sulfate (excess- $\text{SO}_4^{2-}$ ) concentration is remarkably consistent throughout the Arctic. See, for instance, the comparison of Canadian, Norwegian and Alaskan observations by Barrie and Hoff (1984) as well as observations in Greenland by Heidam (1984). Weekly mean excess- $\text{SO}_4^{2-}$  at three locations in the Canadian Arctic for the winters of 1980–1983 (Fig. 7) demonstrate the year-to-year consistency as well as the spatial extent of the haze over thousands of kilometers. Observations from the Arctic aerosol sampling network (Rahn, 1981) together with measurements made in Europe and eastern Canada in conjunction with acid

Table 2. A comparison of estimated anthropogenic trace element emissions in western Europe\*, central Europe† and eastern Europe‡, in 1979 (extracted from Pacyna, 1984)

Element	Emissions ( $\text{ty}^{-1}$ )		
	W	Central	E
As	1600	1320	2810
Be	10	20	15
Cd	868	525	816
Co	499	511	631
Cr	5580	3810	7150
Cu	3390	3370	6540
Mn	5410	3330	6870
Mo	198	267	257
Ni	3830	2920	6010
Pb	39,000	16,900	43,800
Sb	153	104	80
Se	126	111	120
V	9480	5750	11,300
Zn	30,100	14,200	21,300
Zr	403	615	471

\*Western Europe includes Belgium, Denmark, France, the Federal Republic of Germany, Ireland, Netherlands, Norway, Switzerland, the United Kingdom, Luxembourg.

†Central Europe includes Austria, Czechoslovakia, Finland, German Democratic Republic, Hungary, Poland, Sweden, Romania.

‡Eastern Europe is the Soviet Union West of and including the Urals.

rain studies enable one to reconstruct the spatial distribution at ground level of the arithmetic-mean excess- $\text{SO}_4^{2-}$  concentration for the months of January–April. Distributions for 1980 and 1981 are shown in Figs 8 and 9. The isolines are necessarily sketchy due to the lack of observations from the Soviet Union. They are drawn by hand taking into account the configuration of  $\text{SO}_2$  sources (Fig. 4) and the wintertime wind patterns in the northern hemisphere (Fig. 5).

These distributions possess two interesting features:

- (1) There is a 'saddle-point' or minimum in  $\text{SO}_4^{2-}$  concentration located in the N. American–Greenland Arctic between the influence of Eurasian and eastern N. American sulfur sources. This is enhanced by the storminess of the eastern Canadian and Greenland Arctic as indicated by relatively higher precipitation amounts (Fig. 6) in this region.
- (2) Throughout the Arctic and the Eurasian source region,  $\text{SO}_4^{2-}$  concentrations were lower during the winter of 1981 than during 1980. From the time series in Fig. 7, 1981 is 'cleaner' than not only 1980 but also 1982 and 1983 (see also Joranger and Ottar, 1984). Apparently, this is due to very peculiar weather during the winter of 1981. For the northern hemisphere, the period December 1980–February 1981 was the warmest on record since 1881 (Climate Monitor, 1981). Exceptional

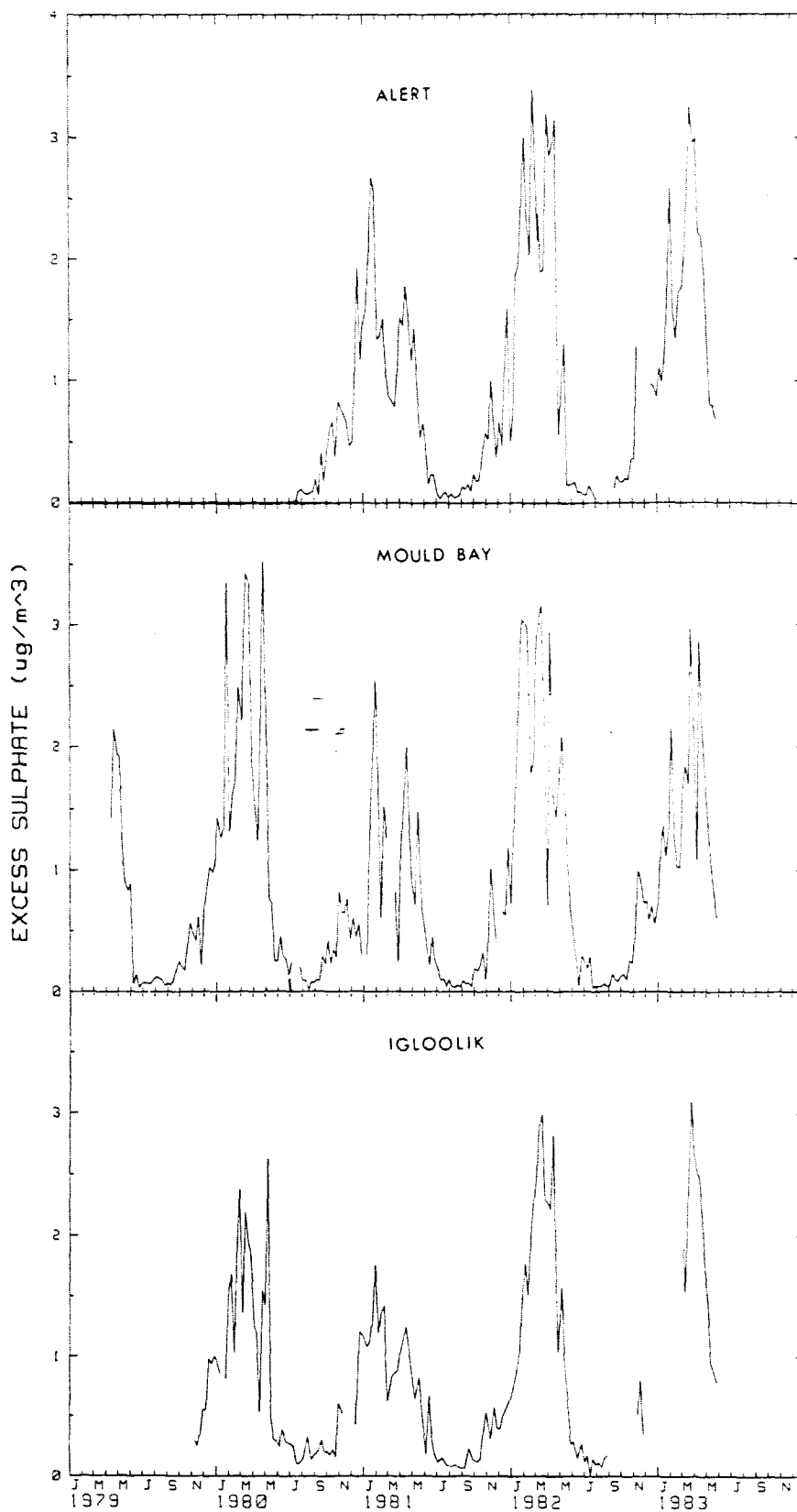


Fig. 7. The temporal variation of weekly mean sulfate in the atmosphere at three locations in the Canadian Arctic (A, M and I in Fig. 1) from Barrie and Hoff (1985).

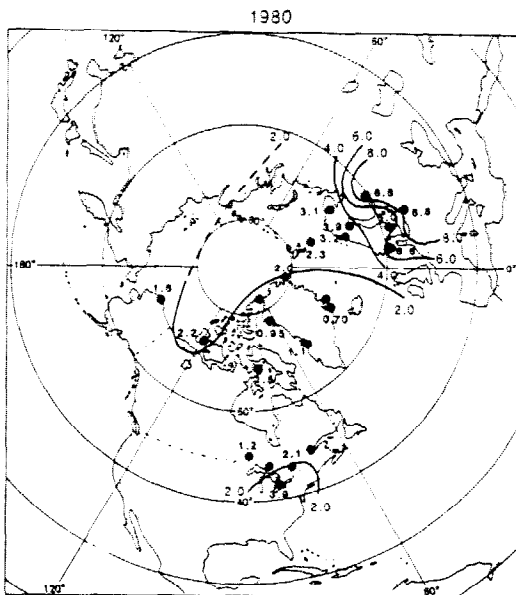


Fig. 8. The spatial distribution of the mean ground-level concentration of sulfate ( $\mu\text{g m}^{-3}$ ) in the atmosphere for the period January–April 1980 (see Fig. 1 for station identification).

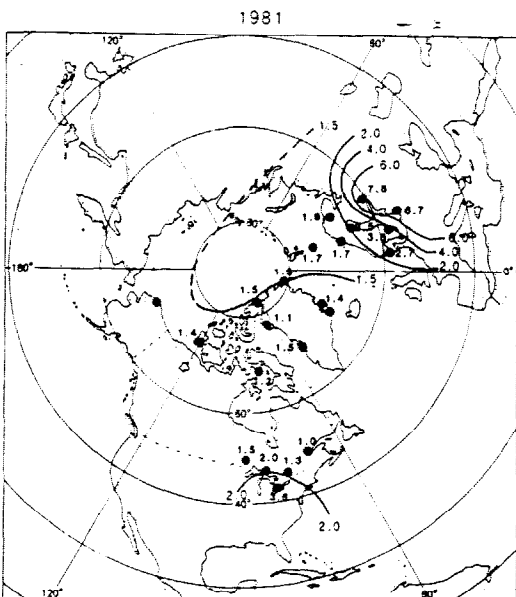


Fig. 9. The spatial distribution of the mean ground-level concentration of sulfate ( $\mu\text{g m}^{-3}$ ) in the atmosphere for the period January–April 1981 (see Fig. 1 for station identification).

warmth in January was particularly marked over the Canadian and Soviet Arctic. Some stations recorded mean temperatures  $15^\circ$  above normal. Thus, the Arctic front did not slip as far S as it normally would. Another factor contributing to lower concentrations is increased ventilation of the source regions caused by abnormally high winds. A

perusal of the general circulation patterns at 700 mb published in *Monthly Weather Review* indicates that average wind speeds over Europe between January and April were higher in 1981 than in 1980.

The vertical distribution of Arctic aerosol is not as well known as the ground-level spatial or temporal distributions. Of the vertical soundings that are published in the literature as of March 1985, 23 are of the aerosol optical scattering parameter  $b_{\text{scat}}$  in dry air (Leaitch *et al.*, 1984; Schnell and Raatz, 1984; Raatz and Schnell, 1984; Radke *et al.*, 1984; Joranger and Ottar, 1984; Hoff and Trivett, 1984), two are of the aerosol optical extinction coefficient  $b_{\text{ext}}$  (Shaw, 1975) and two are of particulate carbon (C) concentration (Hansen and Rosen, 1984). All but one were taken in the spring months March and April.

Since  $b_{\text{scat}}$  (and  $b_{\text{ext}}$  which is approximately equal to  $b_{\text{scat}}$ ) is well correlated with fine particle (accumulation mode) mass (Waggoner and Weiss, 1980) and since most  $\text{SO}_4^{2-}$  and most other anthropogenic particulate constituents are concentrated in the fine particle mode (Hoff *et al.*, 1983; Radke *et al.*, 1984; Pacyna *et al.*, 1984), vertical profiles of  $b_{\text{scat}}$ ,  $b_{\text{ext}}$  and particulate carbon were used to estimate the average vertical distribution of pollution in the Arctic. Each profile was normalized by dividing an observation  $C(H)$  at height  $H$  by the near-surface level value  $C(0)$  [ $C(0)$  refers to  $\sim 1000$  mb] and then calculating the mean and standard deviation of  $C(H)/C(0)$  at levels separated by 500 m. Since some flights were restricted in altitude, the number of normalized profile points available at each height decreased with increasing altitude (22 at 3 km, 16 at 4 km, 7 at 5.5 km and 4 at 7 km). The average profile of particulate pollution and its one-standard-deviation bounds of variability are shown in Fig. 10. The height scale is referenced to the lowest level an aircraft can fly which is about 300 m. Ground-level concentrations are sometimes lower than at 300 m. The particulate mass concentration decreases with height in the first 2 km of the atmosphere to about 50% of the near-surface value. It remains roughly constant up to 5 km. Then between 5 and 7 km, it decreases to about 30% of the near-surface value. The large bounds of variability indicate that at any given time a profile can be very different from the average one. Nevertheless, on average, particulate pollution in spring is found throughout the depth of the Arctic air mass. It tends to be highest in the lowest 2 km.

#### *The size distribution of airborne particles in the Arctic*

The number and volume size distributions of aerosols as well as the mass size distributions of its chemical constituents are information-rich characteristics that provide insight into, among other things, the mechanism of haze formation, the origin of particulate matter and removal rates. Typical number and volume size distributions measured in spring haze near the ground and aloft are shown in Fig. 11. They are based on the



### ARCTIC AEROSOL VERTICAL PROFILE

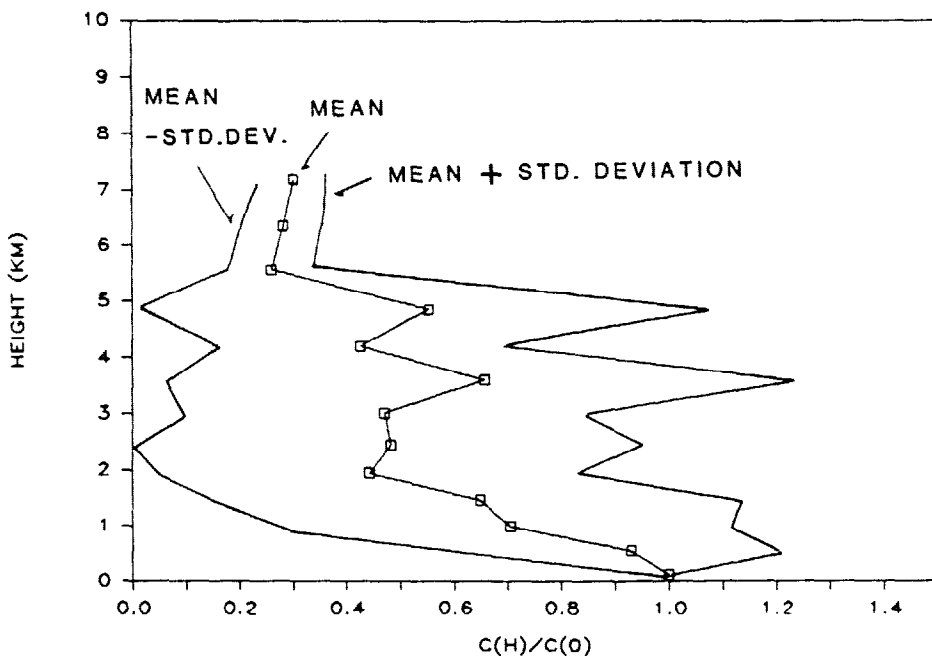


Fig. 10. An estimate of the mean vertical profile of the concentration of anthropogenic aerosol mass in the high Arctic during March and April based mainly on light scattering observations from aircraft (see text).

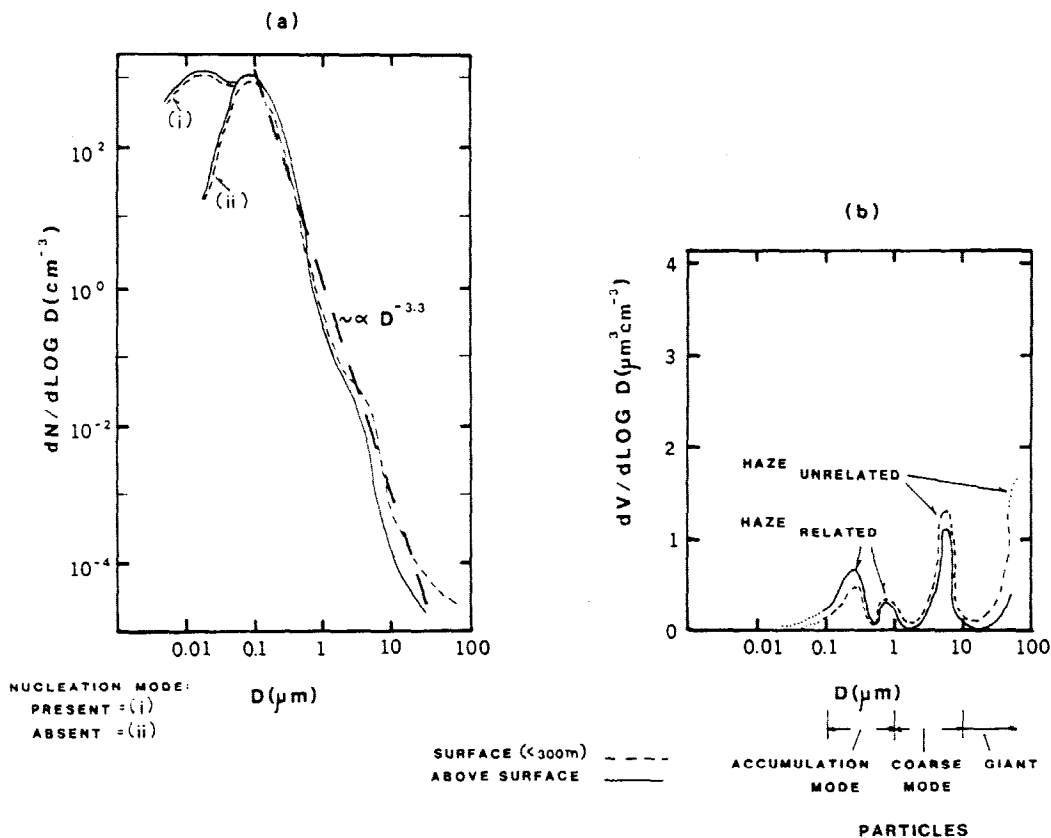


Fig. 11. The number and volume size distribution of a typical Arctic haze aerosol. Adapted from Radke *et al.* (1984), Bigg (1980) and Shaw (1984).

observations of Radke *et al.* (1984) and Shaw (1984) but as discussed below are generally representative of other observations.

Particles are most numerous in the diameter range 0.005–0.2  $\mu\text{m}$ . Above 0.2  $\mu\text{m}$ , their number concentration ( $N$ ) decreases rapidly with increasing diameter ( $D$ ). The functional dependence of  $N$  on  $D$  varies with size range. In the wide range 0.1–30  $\mu\text{m}$ , it depends roughly on  $D^{-3.3}$  [Fig. 11(a)]. In the winter Arctic air mass, the total particle number concentration ( $N_T$ ) ranges from approximately 10 to 4000  $\text{cm}^{-3}$  with a geometric average of 200–350  $\text{cm}^{-3}$  (Heintzenberg, 1980; Bodhaine, 1983; Shaw, 1984, Hogan *et al.*, 1984; Raatz *et al.*, 1985). Hogan *et al.* (1984) report a diurnal variation in  $N_T$  at ground level during February in the Canadian Arctic. It peaked in the day and was a minimum at night. However, it could not be ascertained whether the variation was caused by diurnal variations in boundary layer mixing associated with delivery of particles to the surface from aloft or by *in situ* photochemical production of particles.

The shape of the number size distribution is highly variable below 0.1  $\mu\text{m}$  [Fig. 11(a)]. Sometimes, it decreases much less with decreasing particle size than at other times often showing a second maximum at  $\sim 0.02$   $\mu\text{m}$  (Heintzenberg, 1980; Shaw, 1984; Radke *et al.*, 1984). This is believed to be caused by the production, or lack thereof, of nucleation mode aerosols by condensation of unstable gaseous compounds formed in gas-phase reactions. Radke *et al.* (1984) have evidence that associates nucleation mode particles with sulfur dioxide in the atmosphere and hence presumably the production of sulfuric acid. In more than 80% of the cases in which nucleation mode aerosols were observed,  $\text{SO}_2$  concentrations were greater than 1 ppb(v) while when the concentration was less than 1 ppb(v), no nucleation mode was observed. Natural or anthropogenic organobromides are another possible source of nucleation mode particles. Evidence suggests that in February to April particulate Br forms in the Arctic as a result of photochemical reactions of gaseous organobromides (Berg *et al.*, 1983, 1984). The mass size distribution of Br observed at Igloolik Canada in February (Hoff *et al.*, 1983) supports this conclusion. A large fraction of the Br occurred in particles of aerodynamic diameter less than 0.26  $\mu\text{m}$ . More studies are needed in which the chemical composition of particles less than 0.1  $\mu\text{m}$  are measured simultaneously with number size distributions.

The volume (or mass) of aerosol in the winter Arctic [Fig. 11(b)] is concentrated in larger particles than aerosol number. It is found in three aerosol modes whose formation, optical properties, chemical composition, origin and removal rates differ greatly. These include: (i) the accumulation mode ( $D$ , 0.1–1  $\mu\text{m}$ ); (ii) the coarse particle mode ( $D$ , 1–10  $\mu\text{m}$ ) and (iii) the giant particle mode ( $D > 10$   $\mu\text{m}$ ). Note that some authors define giant particles as those having diameters greater than 1  $\mu\text{m}$  (e.g. Junge, 1963).

(i) *Accumulation mode particles.* Accumulation

mode particles are haze-related because of all particles, they scatter solar radiation and hence reduce visibility most efficiently. They also contain light absorbing soot. In the absence of precipitation, their residence time in the atmosphere is longer than that of other particles because the rate of dry deposition to the earth's surface reaches a minimum between 0.1 and 1  $\mu\text{m}$  diameter (Ibrahim *et al.*, 1983). In the Arctic winter, they are generally anthropogenic in origin. They originate from matter emitted directly to the atmosphere, from the coagulation of nucleation mode particles and from gas-particle interactions. Several investigations have confirmed that particle mass in this size range is concentrated in a distribution centered at  $\sim 0.3$   $\mu\text{m}$  diameter (Bigg 1980; Heintzenberg 1980; Radke *et al.*, 1984). There is some indication of a minor secondary peak at 0.6–0.8  $\mu\text{m}$ . This is suggested by the observations of Bigg and Radke *et al.* in the N. American Arctic.

The detailed study of accumulation mode particle morphology and  $\text{SO}_4^{2-}$  content at Barrow, Alaska using electronmicroscopy by Bigg (1980) resulted in several noteworthy conclusions: (i) winter size distributions are remarkably constant; (ii) sulfuric acid is the dominant winter aerosol. In spring, particles were often more numerous and larger than in winter. Furthermore, they contained a variable and sometimes much greater proportion of ammonium sulfate than in winter; (iii) over half the acid particles contained insoluble inclusions with shapes ranging from spheres through irregular but compact forms to highly irregular aggregates of small particles; the majority of inclusions were single particles; and (iv) the observed size distribution suggests the presence of a sea salt component modified by the addition of sulfuric acid.

(ii) *Coarse particles.* The optically less-active coarse particles originate from soil and to a lesser extent, sea salt. They consist of clay minerals and other soil constituents. Although the number concentration of coarse particles is about four orders of magnitude lower than that of accumulation mode aerosols, they contain a substantial fraction of the aerosol mass. The mass is distributed between 2 and 8  $\mu\text{m}$  with a maximum at 4  $\mu\text{m}$ . The mass concentration is not well correlated with anthropogenic aerosols and haze (Radke *et al.*, 1984).

(iii) *Giant particles.* Giant particles were observed throughout the winter Arctic atmosphere by Radke *et al.* (1984) in sufficient numbers to contain a substantial fraction of total aerosol mass [Fig. 11(b)]. On 10 flights out of Barrow Alaska in April 1983, Bailey *et al.* (1984) observed that, above the surface layer ( $> 0.3$  km) and below 5 km, the number concentration of giant particles was approximately uniform. 10  $\mu\text{m}$  particles occur in concentrations of 100  $\text{m}^{-3}$ . Optical and electron microscopy revealed that the most common major element was iron. Recognizable sea salt particles were rare although particles containing considerable Na and Cl were common. Chain aggregates of soot were also collected regardless of air

mass origin. As a result of particle morphology, the settling velocity of a  $100\ \mu\text{m}$  particle is  $5\ \text{cm s}^{-1}$  which is about a factor of 10 lower than that of a  $100\ \mu\text{m}$  sphere of density  $2.5\ \text{g cm}^{-3}$ . Thus, giant Arctic particles remain airborne much longer than spheres.

In another study of giant particles collected on six AGASP flights in March–April 1983 throughout the Arctic (Winchester *et al.*, 1984), it was concluded that giant particles were soil dust. Two possible sources were suggested. Either the large particles were formed as a result of incorporation into ice crystals which coagulated and subsequently sublimed leaving larger aggregates or they originated from natural emissions from Bennett Island in the eastern Soviet Arctic (EOS, 1983) that occurred in spring 1983. The soil-source explanation is consistent with the observed increase in giant particles in the near surface layer [Fig. 11(b)]. However, more measurements are clearly needed to confirm the existence of giant particles in the Arctic atmosphere and to explain their origin.

The mass size distribution of aerosol trace constituents in the winter Arctic aerosol has been measured in a number of studies (Heintzenberg *et al.*, 1981; Hoff *et al.*, 1983; Pacyna *et al.*, 1984; Radke *et al.*, 1984). The results indicate that most of the total mass of particulate  $\text{NH}_4^+$ ,  $\text{SO}_4^{2-}$ , V, Ni, Cr, Pb, Cu, Br and I is in the accumulation mode range ( $0.1\text{--}1\ \mu\text{m}$ ) while  $\text{NO}_3^-$ ,  $\text{Cl}^-$ ,  $\text{Na}^+$ ,  $\text{Mg}^{2+}$ , Al, Mn, Ca, Ti and Fe have not only a sub-micrometer ( $\mu\text{m}$ ) fraction but also a substantial coarse particle component ( $1\text{--}10\ \mu\text{m}$ ). Pacyna *et al.* (1984) remarked that the presence of Si in the fine particle fraction is evidence that some of the particles in the  $0.1\text{--}1\ \mu\text{m}$  range originate from soil dust. Another explanation is that there is a combustion

source of fine particle Si. They also note that significant amounts of Fe in particles of diameter  $< 0.25\ \mu\text{m}$  diameter suggest that this common soil element is, at least in part, of anthropogenic origin. A hint of this is occasionally seen in the seasonal cycle of aerosol Fe in the Canadian Arctic (Barrie and Hoff, 1985). A maximum in Fe concentration was observed during January and February of 1982 when anthropogenic particulate pollution was at its peak. Concentrations of Ba and P were also observed to peak.

The cation–anion budget for aerosol collected at Igloolik, Canada in February is shown for various particle sizes in Fig. 12 (Hoff *et al.*, 1983). In particles of diameter less than  $1\ \mu\text{m}$ , the dominant soluble ions were  $\text{H}^+$ ,  $\text{NH}_4^+$  and  $\text{SO}_4^{2-}$  while  $\text{NO}_3^-$ ,  $\text{Na}^+$  and  $\text{Mg}^{2+}$  were minor constituents. In particles larger than  $3\ \mu\text{m}$ , the sea salt ions  $\text{Na}^+$ ,  $\text{Mg}^{2+}$  and  $\text{Cl}^-$  were dominant. Between  $1$  and  $3\ \mu\text{m}$ , a mixture of sea salt and acidic sulfates occurred. Thus, the haze-related accumulation mode aerosol is acidic. Barrie *et al.* (1981) found that the equivalents ratio  $\text{H}^+/\text{SO}_4^{2-}$  in Mould Bay aerosols during March 1980 was  $0.6\text{--}0.8$ . On AGASP flights over Alaska, the North Pole and the ocean N of Europe in March/April 1983, the ratio was  $0.41 \pm 0.13$  (Lazrus and Ferek, 1984). These results agree qualitatively with Bigg's (1980) observations discussed above. However, they do not agree with the observations of Cahill and Eldred (1984) who found that the H/S equivalents ratio was  $0.96 \pm 0.05$  in AGASP flight aerosols. These authors point out, however, that ammonium sulfates may have been lost under the vacuum conditions, when they analyzed the sample. On the other hand, inefficient sample handling procedures may have resulted in ammonia contamination

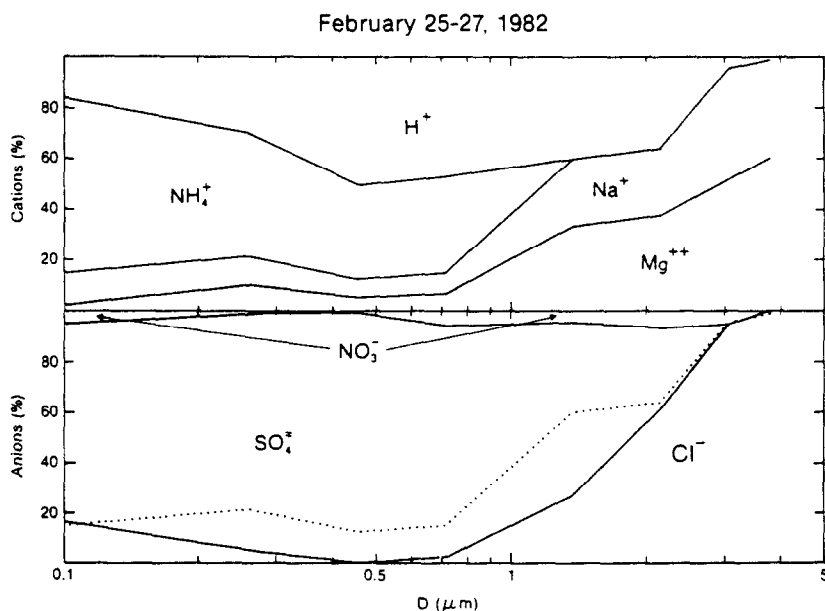


Fig. 12. Cation and anion molar fractions as a function of particle diameter. Hydrogen ion is inferred from an ion balance. The dotted line represents the fraction of anion equivalents required to balance sodium and magnesium ions (from Hoff *et al.*, 1983).

of the samples collected by other investigators. Clearly, more work is needed on the degree of acidity associated with sulfates.

#### *Gaseous pollutants*

Anthropogenic gases play an important role in the Arctic air pollutant scene as precursors of particulate matter, as absorbers of i.r. radiation, agents of biospheric toxification and as tracers of transport pathways to the Arctic. Sulfur dioxide gas is an important source of  $\text{SO}_4^{2-}$  particulate matter. It peaks in concentration in January and February having levels of 0.5–1.5 ppb(v) in the N. American Arctic that increase toward Eurasia. Sulfur dioxide plays a key role in prolonging the occurrence of  $\text{SO}_4^{2-}$  particulate pollution in the spring months March and April well after primary particulate pollution concentrations have peaked (Barrie and Hoff, 1984). Furthermore, there is a strong seasonal variation of  $\text{SO}_2$  oxidation rate in air enroute to the Arctic.

Carbon dioxide gas contributes to the atmospheric greenhouse effect and hence warming. As elsewhere on the globe, its concentration is steadily increasing as a result of man's activities. Concentrations of  $\text{CO}_2$  undergo a stronger seasonal variation in the Arctic than further S at, for instance, Mauna Loa, Hawaii (Peterson *et al.* 1982). The main features of the seasonal variation which are remarkably similar to that of aerosol are caused by biospheric processes of  $\text{CO}_2$  uptake by photosynthesis and release by respiration and biological decay (Pearman and Hyson, 1980). Since in winter biospheric uptake is a minimum in the Arctic air mass,  $\text{CO}_2$  levels rise much higher than over vegetated continents or over oceans. There is direct evidence linking  $\text{CO}_2$  with haze aerosol in the Alaskan Arctic (Peterson *et al.*, 1980). Furthermore, relative maxima in winter  $\text{CO}_2$  concentrations at Barrow and Alert are associated with air mass pathways out of Eurasia (Peterson *et al.*, 1980; Higuchi and Daggupaty, 1985; Halter *et al.*, 1985).

It has been demonstrated by Khalil and Rasmussen (1983) and Rasmussen and Khalil (1984) that anthropogenic organic gases with residence times long enough to ensure transport over 1000s of kilometers but short enough that anthropogenic emissions can be readily observed above global background are useful tracers of transport and possibly as source indicators. Measurements made at Point Barrow, Alaska indicate that  $\text{CHClF}_2$ ,  $\text{CCl}_3\text{F}$ ,  $\text{CH}_2\text{CCl}_3$ ,  $\text{C}_2\text{Cl}_4$ ,  $\text{C}_2\text{HCl}_3$ ,  $\text{CO}$ ,  $\text{CH}_3\text{Cl}$ ,  $\text{C}_2\text{H}_4\text{Br}_2$ ,  $\text{CBrClF}_2$ ,  $\text{CHBrCl}$  and  $\text{CHBr}_2$  fulfill these criteria. These gases have seasonal variations in concentration that parallel that of Arctic haze. The variation is mainly due to seasonal variations in transport rather than in gas-phase chemical destruction mechanisms. Thus, seasonal variations in Arctic haze are caused not only by variations in pollutant removal efficiency but also in transport between mid-latitude sources and the Arctic. This confirms what one would expect from meteorological flow patterns (Fig. 5).

Organic gases can play an important role as i.r. absorbers to enhance the greenhouse effect and as atmospheric tracers. The relationship between various atmospheric trace gases and their concentration-difference between haze and no-haze layers was investigated by Khalil and Rasmussen (1984) in the Arctic during March/April 1983. Gases whose concentrations were well correlated were found to be more abundant within haze layers than outside them. They were generally of anthropogenic origin. They included the high temperature combustion products  $\text{CO}_2$ ,  $\text{CO}$  and  $\text{C}_2$ – $\text{C}_6$  hydrocarbons (HCs) as well as the industrial and commercial chlorofluorocarbons or chlorocarbons (F-11, F-12,  $\text{CCl}_4$ ,  $\text{CH}_2\text{CCl}_3$ , perchloroethylene, trichloroethylene, dichloromethane and chloroform). It is noteworthy that  $\text{CF}_3\text{Br}$  and the solvent F-113 which are used in the high technology electronics industry of the United States were not observed.

On the basis of measurements at Spitsbergen in the Norwegian Arctic, Hov *et al.* (1984) concluded that, with the exception of ethene and propene, the concentrations of halocarbons and light HCs were an order of magnitude higher in March 1983 than in July 1982. It was also concluded that air reaching Spitsbergen from an easterly direction (i.e. the central Soviet Union) is much richer in alkanes (presumably from oil and gas combustion), much less abundant in chlorinated ethenes and of comparable abundance in benzene and toluene than air sampled at Barrow.

The long-range transport of pesticides to the Norwegian Arctic has been documented by Oehme and Ottar (1984). Consequently, the accumulation of polychlorinated HCs in Arctic ecosystems can be expected. Measurements of the pesticide chlordane at Mould Bay, Canada, in June and July 1984 (Hoff and Chan, 1986) confirm the existence of this potentially toxic man-made substance in an area very remote from sources. They indicate that the atmospheric pathway may be important in explaining the levels of such chemicals found in fish and mammals of the N. American Arctic (Muir *et al.*, 1986). More measurements are needed of such airborne organic compounds to establish seasonal variations.

#### DEPOSITION OF POLLUTANTS: PRECIPITATION AND GLACIER EVIDENCE.

Deposition of pollutants to Arctic ecosystems is affected by precipitation scavenging (wet deposition) and by direct uptake at the earth's surface (dry deposition). The processes involved are discussed in detail by Barrie (1985). There are several ways of obtaining information on deposition.

- (1) Measure air concentrations of pollutants and estimate deposition based on parameterizations of the scavenging processes. For instance, Barrie *et al.* (1981) used aerosol acidity measurements at Mould Bay and a volumetric scavenging ratio of 2

$\times 10^5$  to predict the seasonal variation of precipitation acidity in the Arctic. A similar variation in dry deposition of acidity was expected. These predictions were later confirmed by glacial ice core measurements (Koerner and Fisher, 1982; Barrie *et al.*, 1985).

- (2) A more direct way of obtaining wet deposition estimates is to analyze precipitation samples. To the author's knowledge, there are at present no well-quality-controlled precipitation chemistry measurements being made on a routine basis in the Arctic. Precipitation amount in the region during winter is very low (typically 5 mm water equivalent per month) and blowing snow is a serious practical problem. This together with relatively low pollutant concentrations makes quality control of measurements even more essential. There have been several reports of precipitation chemistry measurements in the Arctic during summer. McNeely (1982) found the pH of precipitation on Ellesmere Island, Canada in June and July 1981, to be  $5.7 \pm 0.5$ . At Poker Flats, Alaska, 16 samples of precipitation collected in December–January 1979 and in May–December 1980 had a mean pH of 5.0 that was caused mainly by sulfuric acid (Galloway *et al.*, 1982). Dayan *et al.* (1985) have examined these and more measurements from Alaska in summer and confirmed the dominance of sulfuric acid. Formic and acetic acids contributed a maximum of 33% to the free acidity. However, few measurements were available for the winter period when Arctic air pollution is at its worst. A snowpack chemistry survey on central Ellesmere Island between May 1979 and August 1981 was undertaken by McNeely and Gummer (1984). Measureable concentrations of several pesticides were observed (Table 3). Lindane and its isomer  $\alpha$ -BHC were most abundant.
- (3) Another way to obtain information on deposition is to collect snowpack samples. Provided care is taken that no leaching of the sample by meltwater during a melt has occurred, the composition of the pack will be determined by atmospheric inputs and soil constituents. If air measurements are conducted simultaneously, information on the relative

magnitude of wet and dry deposition can be obtained (Davidson *et al.*, 1981, 1985).

A snowpack chemistry survey in the Soviet Union during the spring of 1982 (Belikova *et al.*, 1984) yielded the spatial distribution of sulfate concentration shown in Fig. 13. The results are consistent with the configuration of  $\text{SO}_2$  emissions in Eurasia (Fig. 4). Concentrations are highest in the western Soviet Union, around Norilsk, in eastern Siberia and in the S central region.

A snowpack chemistry survey on Spitsbergen in spring 1983 (Semb *et al.*, 1984) yielded mean concentrations for  $\text{H}^+$ ,  $\text{NH}_4^+$ ,  $\text{NO}_3^-$  and non-sea salt  $\text{SO}_4^{2-}$  in the top 30 cm of snow of 12, <1, <1 and  $12 \mu\text{eq l}^{-1}$ , respectively. Where comparison was possible, results agreed reasonably well with earlier measurements made by Gjessing (1977). Note that most of the acidity was associated with the  $\text{SO}_4^{2-}$  ion and that the pH of snow was 4.9. In a snowpack chemistry study in northern Alberta conducted by Barrie (1980), it was found that even at locations remote from the influence of local emissions, the snowpack during March had an  $\text{H}^+$  concentration of  $8\text{--}20 \mu\text{eq l}^{-1}$  (pH 4.7–5.1) and the predominant anions were  $\text{SO}_4^{2-}$  and  $\text{NO}_3^-$ . This is considerably more acidic than summer precipitation. Another snowpack chemistry survey conducted in the Northwest Territories of Canada in March 1984 (Shewchuk, 1986) yielded, at sites on the pre-Cambrian shield remote from calcareous soils,  $\text{H}^+$ ,  $\text{NH}_4^+$ ,  $\text{NO}_3^-$  and  $\text{SO}_4^{2-}$  concentrations in the range 2–10, 1–2, 5–7 and 7–18  $\mu\text{eq l}^{-1}$ , respectively. Thus, the snowpack is slightly acidic and there is sufficient  $\text{SO}_4^{2-}$  and  $\text{NO}_3^-$  to explain the acidity.

The concentration of various trace elements in the winter snowpack of northern Alaska was investigated by Weiss *et al.* (1978) and by Rahn and McCaffrey (1979b). The results are compared in Table 4. Enrichment factors relative to crustal rock are also shown. Concentrations of all elements compare well. In addition, the snowpack is so highly enriched in Zn, Cd and Hg relative to crustal material that there is little doubt that they are mostly of anthropogenic origin. Elements with

Table 3. A summary of the concentrations ( $\mu\text{g l}^{-1}$ ) of man-made pesticides in the snowpack of central Ellesmere Island in May–August of 1979–1981 (McNeely and Gummer, 1984)

Pesticide	Number of analyses	Number of samples above detection limit	Concentrations	
			Min	Max
$\alpha$ -BHC	28	21	<0.001	0.018
$\gamma$ -BHC (lindane)	28	10	<0.001	0.008
$\alpha$ -Chlordane	28	1	<0.001	0.002
$\gamma$ -Chlordane	28	2	<0.002	0.002
$\alpha$ -Endosulfan	28	1	<0.001	0.002
op' DDT	28	2	<0.001	0.002
pp' DDT	28	2	<0.001	0.002
HEOD (dieldrin)	28	5	<0.002	0.004

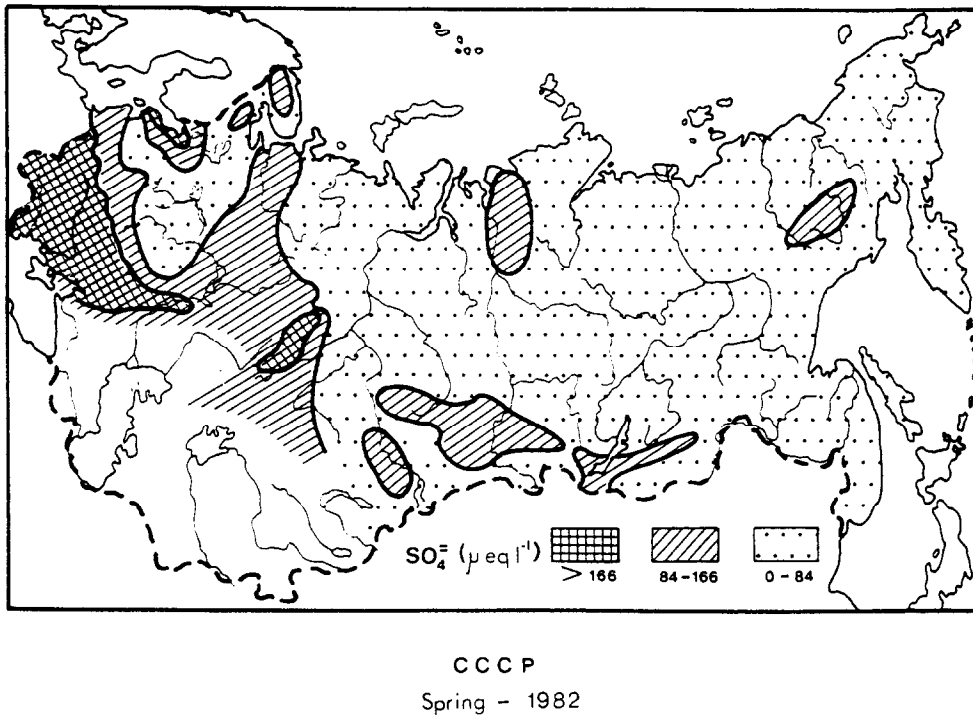


Fig. 13. The spatial distribution of snowpack sulfate concentration in the Soviet Union during spring 1982 (from Belikova *et al.*, 1984).

Table 4. The concentration ( $C$ ) and crustal enrichment factors ( $EF$ ) of trace elements in the winter snowpack on the north slope of Alaska near Barrow.

Element	Weiss <i>et al.</i> (1978)		Rahn and McCaffrey (1979b)	
	$C^*$ ( $\mu\text{g l}^{-1}$ )	$EF^\dagger$	$C^\ddagger$ ( $\mu\text{g l}^{-1}$ )	$EF^\ddagger$
Na	104	7.6	144	15.9
Mg	50	5.0	41	6.1
Al	39	1.0	26	1.0
Ca	61	3.5	100	8.6
V	0.109	1.7	0.086	2.0
Mn	0.99	2.2	0.79	2.6
Zn	1.12	33	1.16	52
Cd	0.07	724	0.047	730
Hg	<0.05-0.014	—	0.005-0.020	~430

\*The arithmetic mean of all samples in Table 1 of Weiss *et al.* (1978).

†Relative to crustal aluminum as given by Rahn and McCaffrey (1979b).

‡From Table 1 of Rahn and McCaffrey (1979b).

moderate enrichments such as Na and Mg are likely of marine origin. The enrichment for Ca is likely a characteristic of Ca rich soil on the N slope. In a comparison of enrichment factors in the snowpack with those in the Arctic haze aerosol, Rahn and McCaffrey (1979b) concluded that relative to crustal material Na, Mg, V, Zn, Cd and Hg were more enriched in the aerosol than in the snow. This is probably due to the large amounts of soil material in the snow pack originating from wind erosion of local outcrops. This debris is usually in such large particles that it does not get into the atmospheric aerosol. Another contributing factor

is the higher deposition rate of soil-derived particles over smaller anthropogenic particles.

- (4) Another important source of information on the deposition of Arctic air pollution is glacial ice. Measurements of snow and ice composition have been made in Greenland (Busenberg and Langway, 1979; Hammer *et al.*, 1980; Herron, 1982; Davidson *et al.*, 1981, 1985), on northern Ellesmere Island, Canada (Koerner and Fisher, 1982; Barrie *et al.*, 1985) and on Mt. Logan in northwestern Canada (Holdsworth and Peake, 1984). The elevation and location of these sampling sites are important factors in interpreting the observations. For inst-

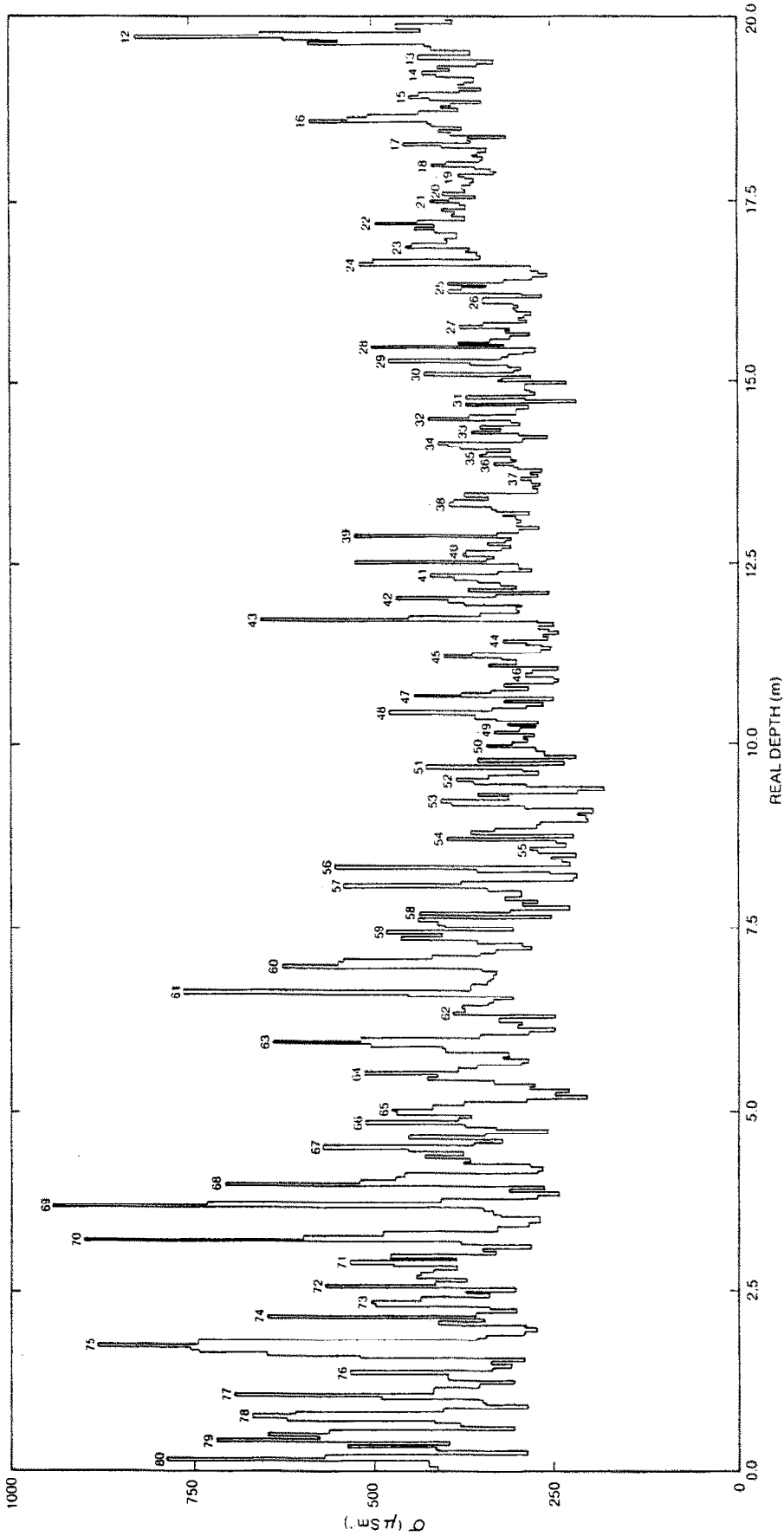


Fig. 14. The variation of melt-water conductivity in the Agassiz glacier, Ellesmere Island, with depth (location near A in Fig. 1), from Barrie *et al.* (1985).

ance, in going from northern Ellesmere Island to Greenland to Mt. Logan elevation changes from 1.6 to 2.5 to 5.3 km. Considering the vertical profile of Arctic air pollution (Fig. 10), Mt. Logan and Greenland glaciers are receiving snow from relatively unpolluted middle and upper tropospheric air while Ellesmere Island glaciers are sampling polluted lower tropospheric air. Tropospheric folding events such as that described by Shapiro *et al.* (1984) over Greenland can bring stratospheric air as low as 3 km into the troposphere.

The conductivity and acidity of snow on the Agassiz glacier on northern Ellesmere Island are well correlated and, as shown in Fig. 14, undergo a strong seasonal variation paralleling that of Arctic air pollution (Barrie *et al.*, 1985). The maximum conductivity (acidity) in a year occurs at the peak of Arctic air pollution and is therefore an indicator of the levels of air pollution. Indeed a comparison of the historical record of maximum ice core conductivity (acidity) with that of  $\text{SO}_2$  emissions in Europe (including the western Soviet Union) during this century supports this hypothesis (Fig. 15). As emissions remained roughly constant in the first part of the century, ice core conductivity remained roughly constant with the exception of a peak in 1912–1913 associated with the eruption of Mt. Katmai in the Aleutian Islands in 1912. Then between 1956 and 1977, as  $\text{SO}_2$  emissions in Europe doubled, ice core conductivity underwent a

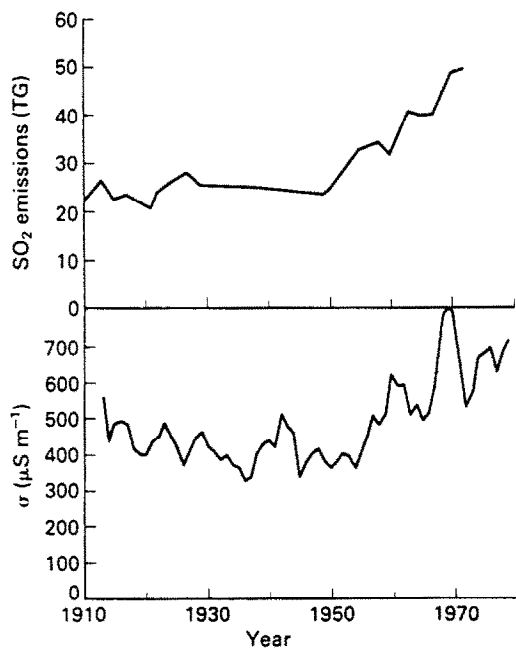


Fig. 15. A comparison of the historical record of annual maximum conductivity in melt-water from the Agassiz glacier, Ellesmere Island (data in Fig. 15) and  $\text{SO}_2$  emissions ( $10^6 \text{ Mt y}^{-1}$ ) from Europe (including the western Soviet Union) from Barrie *et al.*, 1985. The conductivity record is a three-year running mean.

marked increase by 75%. Current levels of winter time acidity are  $12\text{--}14 \mu\text{eq l}^{-1}$  compared to  $7\text{--}9 \mu\text{eq l}^{-1}$  prior to 1956. These levels of acidity are quite comparable to the  $12 \mu\text{eq l}^{-1}$  observed in Spitsbergen snow by Semb *et al.* (1984) and to the  $2\text{--}10 \mu\text{eq l}^{-1}$  observed in central Canada by Barrie (1980) and Shewchuk (1986). Limited historical records of aerosol optical depth in the Arctic atmosphere in March do not contradict the general trend suggested by the Agassiz ice cap data (Shaw, 1982).

How do historical records of glacial acidity on Greenland and Mt. Logan compare with those obtained on Ellesmere Island? A historical record on Greenland and reported for Dye 3 by Herron (1982) is shown in Fig. 16. Although acidity was not measured, the concentrations of the acid-related ions  $\text{SO}_4^{2-}$  and  $\text{NO}_3^-$  were measured. The results indicate that there has been an increase in concentrations in this century; for  $\text{SO}_4^{2-}$  an increase of about  $1 \mu\text{eq l}^{-1}$  to current levels of  $1.7 \mu\text{eq l}^{-1}$  and for  $\text{NO}_3^-$  an increase of 1.3 to  $2.0 \mu\text{eq l}^{-1}$  currently. Recent measurements of  $\text{SO}_4^{2-}$  and  $\text{NO}_3^-$  concentration trends in this century on S. Greenland (Nefel *et al.*, 1985) confirm Herron's observations. There has been a significant increase since the late 1930s. Ice core

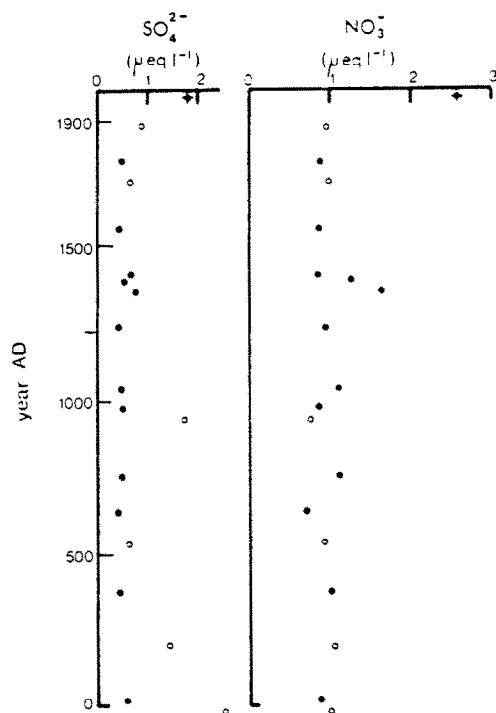


Fig. 16. Historical records of  $\text{SO}_4^{2-}$  and  $\text{NO}_3^-$  concentration from the Greenland ice cap at Dye 3 (extracted from Herron, 1982). Open circles denote intervals with volcanic impurities and solid circles show periods without. Each point is a multi-year average over a period 2–37 y. The sample in this century is emphasized with a cross.



acidity at another location on Greenland was found to display no trend between 1850 and 1950 (Hammer *et al.*, 1950). Thus, most of the increase in this century took place since 1950. Further support for this is found in historical records of Pb in Greenland ice which indicate that levels have risen since about 1950 (Boutron, 1980). This is consistent with observations lower in the troposphere on Ellesmere Island (Fig. 15).

Apparently, Greenland which is sampling middle tropospheric air has seen as great an increase in  $\text{NO}_3^-$  as in  $\text{SO}_4^{2-}$ . Whether this is the case on Ellesmere Island down in the more polluted lower troposphere remains to be seen. Since, on an equivalents basis, ground-level aerosol  $\text{SO}_4^{2-}$  is much more abundant in the Arctic than aerosol  $\text{NO}_3^-$  plus  $\text{HNO}_3$  (Barrie *et al.* 1981; Barrie and Hoff, 1985), it is reasonable to expect a greater abundance of  $\text{SO}_4^{2-}$  than  $\text{NO}_3^-$  in Ellesmere Island snow.

Measurements of pH in the glacial ice of Mt. Logan yield a mean value of 5.5 for 1979–1981 (Holdsworth and Peake, 1984). There is no apparent trend in acidity in this century like that found on Ellesmere Island. This is to be expected since Logan is at 5.3 km altitude adjacent to the climatologically persistent Aleutian low pressure system in the N. Pacific (Fig. 5). Consequently, it sees upper tropospheric air off the Pacific Ocean rather than Arctic air pollution. The mean concentration of  $\text{NO}_3^-$  and non-sea salt  $\text{SO}_4^{2-}$  is 0.8 and 0.5–0.9  $\mu\text{eq l}^{-1}$ , respectively.

#### THE IMPLICATIONS AND EFFECTS OF ARCTIC AIR POLLUTION

Arctic air pollution is widespread and covers large parts of the northern hemisphere during the winter period of January–April. However, is this pollution really cause for concern? What are the implications of Arctic haze? Are there serious environmental effects? These are legitimate questions that must be answered by the scientific community *before* alarm bells are rung. One important implication of the Arctic haze phenomenon is that during the winter half of the year the Arctic air mass is more 'pollutable' than any other in the northern hemisphere. In other words, pollutants are more inefficiently dispersed and removed in the cold northern atmosphere than elsewhere. This fact should be taken into account in planning future industrial development of the N. There are several real or potential effects of Arctic air pollution that warrant attention at this time:

##### (1) Reduction of visibility

Arctic particulate matter scatters solar radiation thereby making distant objects more obscure and reducing visibility. Using the Koschmieder relationship between visibility and the optical extinction

coefficient as well as the correlation between aerosol scattering in dry air ( $b_{\text{scat}}$ ) and  $\text{SO}_4^{2-}$  aerosol concentration in  $\mu\text{g m}^{-3}$  (Barrie *et al.*, 1981; Barrie and Hoff, 1985), it can be shown that dry aerosol visibility ( $V$  in km) is given by:

$$V = \frac{3910}{11[\text{SO}_4^{2-}] + 6}. \quad (1)$$

Thus, for  $\text{SO}_4^{2-}$  concentrations typically found in the winter Arctic of 1–4  $\mu\text{g m}^{-3}$ , one can expect visibilities in low relative humidity situations to be 230–78 km. Actually, in the Arctic relative humidity is high. Consequently, these dry air visibilities are overestimates. Even if they are high by a factor of 2, on average, one cannot obtain a reduction of visibility of less than 30 km from anthropogenic aerosols alone. However, visibilities of 10 km are often observed in the winter Arctic even in the absence of blowing snow (Leitch *et al.*, 1984). Most likely this is caused by the presence of ice crystals in the atmosphere which often accompany haze aerosols. The physical–chemical link between aerosols and ice crystals needs to be investigated on a more quantitative basis. If there is indeed a link as current evidence suggests (Otake, 1984), then reduced visibilities are indirectly caused by anthropogenic aerosols. It seems that below  $-25^\circ\text{C}$ , ice can be nucleated in acidic accumulation-mode aerosols which then grow into ice crystals. These crystals may drop from the atmosphere carrying pollutants with them.

##### (2) Climatic effects

The presence of soot-bearing anthropogenic particulate matter in the Arctic can affect the energy budget of the atmosphere and hence, potentially influence climate. The energy budget is influenced in two ways. First, as a black aerosol suspended over a white surface, pollution increases the net amount of solar radiation trapped in the troposphere. For instance, based on optical measurements on one flight in March 1983 in the N. American Arctic, an increase of 36% in the total absorbed solar radiative energy was associated with haze (Valero *et al.*, 1984). Second, soot-laden aerosols increase the blackness of the snowpack when they are deposited. Hence, the amount of sunlight trapped is increased. Studies by Clarke and Noone (1985) indicate that the albedo is decreased by 1–3% in new snow and by 3–10% in old snow. This effect can last well into May and June even after airborne anthropogenic pollution has disappeared. During spring melt, the blackness of the snowpack increases as water but not soot runs off leaving black soot behind.

The net result of the above perturbations to the solar radiation budget is the potential modification of local climate and possibly the hemispheric climate. Attempts to model the perturbation to the solar radiation budget have been made using radiative transfer models (Shaw and Stamnes, 1980; Porph and McCracken, 1982; Cess,

1983). However, as pointed out by Cess, they suffer from numerous drawbacks, one of which is the assumption of clear skies. Furthermore, they are not climate models that link atmospheric dynamic processes and climate feed-back mechanisms with the perturbation. Arctic aerosols need to be incorporated into global climate models as the next step in assessing the impact of Arctic air pollution on climate. A preliminary attempt to examine the effects of aerosols on a global basis (Cess and Potter, 1984) led to the conclusion that particulate matter reduces global-mean surface temperature by 3–4°C and that maximum cooling occurs at high latitudes despite the fact that particles produce an increase in solar heating at these latitudes. However, this model did not take into account the high absorptivity of black Arctic aerosols or the decrease in albedo of the snowpack as discussed above or seasonal variations in the aerosol.

### (3) Pollutant deposition effects

As mentioned in the previous section observations of deposition indicate that snow falling in the winter Arctic is slightly acidic. pH ranges from 4.9 to 5.2. To this author's knowledge, analyses of the effects of acidic snow on delicate ecosystems in the Arctic have not been published. There is some concern that during spring melt, selective leaching of  $H^+$  ions from the snowpack by the first melt-water can lead to a three- to five-fold concentration of acidity and an acid shock (Johannessen and Henriksen, 1978; Jeffries *et al.*, 1979). Runoff is not likely to be acidic unless the soil is poorly buffered. There are large areas in the Canadian

Arctic that have poorly buffered soils (Fig. 17). Thus, until biologists assess the impact of an acidic runoff at the beginning of spring melt in these areas little can be said about the importance of this effect.

A potentially more serious effect is the deposition and accumulation of man-made pesticides transported from mid-latitudes into the Arctic. As noted earlier, lindane has been observed in the snowpack of Ellesmere Island (McNeely and Gummer, 1984). However, once again until the effects of such deposition are assessed by biological research we must 'hold our breath' and wait.

Few budgets of organics and trace elements in Arctic ecosystems have been calculated to assess the importance of the atmosphere as a source. One notable exception was a budget estimate for the Arctic Ocean done by Rahn (1981) for Al, V, Mn, Cd, Pb,  $SO_4^{2-}$  and  $NO_3^-$ . It was concluded that Pb was the only element whose atmospheric source equalled or exceeded riverine and oceanic sources. More such studies are needed for pesticides.

### CONCLUSION

The Arctic atmosphere is not a pristine environment untouched by man. In the winter period of December to May, elevated levels of man-made particulate matter originating mainly from mid-latitudinal sources in Eurasia are found throughout the Arctic. For the rest of the year, particulate pollution is either absent, or present at much lower concentrations than in winter.



Fig. 17. Areas potentially sensitive to acid precipitation in northern Canada (first approximation) from Rubec (1981).

This overview of current knowledge shows that, in the last 10 years, we have come a long way towards understanding the occurrence, nature, history and origin of Arctic pollution. However, much remains to be learned particularly in describing the occurrence of pollution in the Soviet Arctic, in understanding the interaction between particulate pollution and the water vapor cycle and in assessing the effects of pollution on visibility, climate and terrestrial-aquatic ecosystems. Furthermore, more measurements of pollutant vertical profiles are needed. Another important area for future applied research is the development of chemical-transport models for the northern hemisphere poleward of 30°N. These models will be useful assessment tools in the planning of future industrial development in the north. Models in conjunction with chemical tracer techniques involving aerosols and gases have the potential to more quantitatively define the relative contributions of Arctic pollution sources.

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