Vertical Structure of Arctic Haze Observed by Lidar

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(Manuscript received 20 February 1987, in final form 2 July 1987)

ABSTRACT

A Mie scattering lidar was operated at Alert, NWT, Canada, for 9 weeks for the winter of 1984/85 in order to determine vertical profiles of Arctic haze. During the study period, the strong sulphate aerosol concentration maximum, representative of previous years, was not found due to a low pressure system which remained in the Baffin Bay for much of the winter. The relatively clean air is believed to be typical of southerly air flow to Alert. The vast majority of lidar profiles, which did not contain scattering from hydrometeors, showed little change in scattering structure with height. Several events of increased light scattering due to aerosols were noted and allowed examination of three possible mechanisms for the vertical movement of haze: motions following potential temperature isentropes, foehn (chinook) development and ice crystal precipitation scavenging.

1. Introduction

In the study of the Arctic haze phenomenon, one of the most elusive parameters and one which is crucial to the understanding of aerosol transport climatology is the vertical structure of the haze aerosol. Previous studies of the Arctic aerosol and light scattering profile have drawn the representativeness of surface observations of Arctic haze into question (Hansen and Rosen, 1984; Bodhaine et al., 1984; Schnell and Raatz, 1984). One result (Hansen and Rosen, 1984) has shown that the surface carbonaceous aerosol concentration is one-half that at the top of the inversion. Due to the strongly stratified nature of the Arctic troposphere, it is necessary to define the mechanisms which move aerosol to the surface from the transport altitudes of the lower troposphere. Strong inversions limit vertical mixing in winter and it may be inappropriate to interpret events with high ground concentrations by assuming that the aerosol has conserved this high concentration along an isobaric 1000 mb back trajectory. If the material were transported above the inversion and mixed downward by some relatively local mechanism, completely different source regions might be indicated.

As early as 1950, Greenaway (1950) reported ice crystal hazes to 6.7 km and, in a now classic report, Mitchell (1957) suggested that haze in the Arctic was less than two microns in size and could be found up to a 9 km altitude. Past studies using instrumented aircraft (Hoff et al., 1983; AGASP, 1984; Ottar et al., 1986) have provided a very limited picture of the climatology of haze structure. In some 27 aircraft profiles of aerosol concentration (inferred from light scattering and particulate carbon measurements) taken in the Arctic and published before the winter of 1984/85, Barrie (1986) has summarized the results to show that the Arctic aerosol is generally most intense at the surface with a 50% drop-off in concentration by 2 km altitude. Flights made during the Arctic Gas and Aerosol Sampling Program (AGASP I and II) showed that layers of haze might be present to altitudes of 6 km (Schnell and Raatz, 1984).

In order to ascertain the representativeness of the haze profiles previously obtained, a monostatic ruby lidar was installed at Alert (82°30'N, 62°18'W) from September 1984 to March 1985. Use of a vertically pointing lidar has been proven to be effective in routinely monitoring aerosol light scattering coefficients in urban and rural areas in the midlatitudes (see for example, Collis and Russell, 1976) and has been used at Global Monitoring for Climate Change (GMCC) sites at Mauna Loa (DeLuisi et al., 1986) and the South Pole (Smiley and Morley, 1981).

As a remote sensor, the lidar observes atmospheric aerosols as they appear in nature, unmodified by sampling effects. As such, the results obtained reflect more realistically the light scattering characteristics of the in situ aerosol than those obtained by integrating nephelometers, for example, which heat the aerosol and dry it before measurement. This proved to be extremely useful during the measurements of the winter of 1984/85 in identifying the causes of severely reduced atmospheric visibilities. As will become obvious later, light scattering from large ice crystals often dominated the scattering from the smaller (anthropogenic) haze aerosols.

Six separate weekly studies were conducted between September 1984 and February 1985. The data from a

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3-week continuous duration study of March 1985 will be presented elsewhere; however, a few results of that study will be included here. While the project design was clearly expected to give information on the vertical profile of pollutant aerosols, Arctic meteorology often confuses both the instrument and the observer. Results will be discussed in the following sections in terms of vertical aerosol structure, ice crystal light scattering, ice crystal precipitation mechanisms, and the meteorological implications of the results in terms of haze transport.

2. Experimental

The ruby lidar used in this study had previously been used for monitoring dispersion of industrial plumes (Hoff and Froude, 1979). With this lidar, a pulse of intense red light (694.3 nm at 0.5 J output energy) was transmitted vertically through an evacuated tube in the roof of a building at Alert. Extreme care was necessary to prevent frosting of the transmitter and receiver windows in the Arctic winter cold, and, in addition to the evacuated transfer tubes, heating of the pyrex exterior windows was required. The receiver consisted of a 20 cm diameter Fresnel telescope, neutral density and polarizing filters, an RCA C31000A photomultiplier, an Analog Modules LA-90-P logarithmic amplifier and a LeCroy TR8827 32 MHz digitizer. Data were recorded on a Compaq Plus portable PC.

Lidar pulses were fired on 2-min intervals during this study. The lidar equation was solved for the backscattering coefficient of the aerosol, assuming no two-way transmission losses in the signal (an excellent assumption for much of the low extinction situations encountered in the Arctic when only anthropogenic aerosol is present). After output power normalization and range scaling of the return signal, color displays of the time–height lidar profiles of the Rayleigh ratio (R) were prepared. Here R is the ratio of the backscattered power (proportional to the backscatter coefficient of the aerosol) to the backscattered power expected from a clear air (Rayleigh) atmosphere at STP. No correction was made for change of pressure with height and the maximum deviation of R from the true Rayleigh backscattering ratio at the proper temperature and pressure was 20% for the study period. Minimum vertical resolution of the lidar was 4.6 m with a range of 2400 m. Because of the large dynamic range difference in returned signals, it was common to lose information on the lowest 200–300 m of the atmosphere in order to obtain adequate signal sensitivity at the top of the profile. Cloud returns were obtained to 12 km. In clear air, a combination of instrumental effects and low aerosol content typically limited useful sensitivity to less than 3 km in altitude.

On a manual basis, polarization diversity was used to discriminate against nonspherical scatterers. In this mode, a crossed polarizer was added to the receiver chain and only the perpendicular component of the scattering (P⊥) detected. The depolarization ratio

\[ D = P_1(P_\perp + P_i)^{-1} \]

is related to the irregularity of the particle and crystal type for ice crystals (Sassen, 1977). In this study, \( D < 0.1 \) was used to indicate that the scatterers were anthropogenic aerosols or gases. Since the manual intervention of the polarizer was irregular, it was used primarily as a check on the general features of the scattering aloft.

In addition to the lidar data, Alert has a rawinsonde station which provides both hourly surface meteorological observations and upper air profiles on a 12 h basis. These results allow direct comparison with the lidar data.

3. Vertical aerosol profiles

The derived values of R have been examined as time–height diagrams and time series. Figure 1 shows a time–height color intensity display from the 19–20 November 1984 sampling period. A well-defined layer of about 100 m thickness was seen at an altitude of 600 m above the surface at 1200 UTC and descended to the surface by 1200 UTC on the 21st. The dark bands in the figure are periods when the cross-polarization component of the return was examined. D was found to be less than 0.1 for the lower layer but not for the cloud seen at 2100 UTC at 1800 m. For the cloud, either ice crystal (nonisotropic) or multiple scattering is the reason for the loss of coherency in the polarization.

Time series of R in 140 m layers above 140 m MSL and less than 2240 m MSL were also examined. The time series proved to be less useful since comparison of R to other observables is often not straightforward. Time series of visibility obtained from R will be discussed below.

In most of the weekly studies, it was practically impossible to obtain a completely continuous scattering record. The main reason for this was the relatively common occurrence of snow, ice crystals or ice fog in the valley where the lidar was located. During the winter months, ice fog can be expected when the temperature falls below \(-39^\circ\text{C}\). Fortunately for the Alert site, the long-term WMO BAPMON aerosol sampling site is on a remote plateau some 200 m above the main camp. This altitude both decouples the local camp pollution (including H\textsubscript{2}O pollution) and provides a warmer location for the sampler. During the most severe cases of camp fog seen during the 1984/85 winter, the problem seemed well confined to the valley in which the lidar was located. (This did not make life any more enjoyable for the operators, however.) As an estimate, about one-third of the operating time was free of poor weather, ice crystals and fog so that anthropogenic aerosol could be monitored unambiguously. All of the
Fig. 1. Time–height color display from 1200 UTC on 19 November 1984, to 0000 UTC on 21 November. The color intensity is proportional to the Rayleigh ratio.

Fig. 4. Time–height color display of the 6 March event showing the aerosol following isentropic surfaces.
aerosol data in the study was obtained when there was a wind blowing the camp plume away from the lidar site.

Sulphate aerosol concentrations measured by the high volume sampler in 1984/85 were not typical of previous years. Concentrations of SO₄²⁻ at Alert were abnormally low from November to early April. Figure 2 shows the weekly averaged sulphate concentration over the winter and it is noted that the sulphate concentrations over this period are the lowest for this site since the winter of 1981. The primary reason for this is the stagnation of a polar low in Baffin Bay after October, which locked Alert into a generally southerly flow for much of the winter. Since high aerosol concentrations are most often correlated with polar trajectories (Barrie and Hoff, 1985), the lower aerosol concentrations from the Canadian Arctic were to be expected.

Over the course of the winter, when ice crystals, ice fog, and cloud were not present, R ranged from 1 to about 7. Figure 3 shows the average R profiles for the weekly samples of November to February. Clearly, there is an increase in light scattering as the spring Arctic haze maxima approaches. Except for the January study which shows a 50% increase in light scattering aloft, the average profiles are generally uniform with height.

Since higher levels of sulphate aloft may well be correlated with warmer, moister air masses (from southerly transport), the lidar measurements may have missed these events due to obscuration by cloud or ice crystals. Discrimination of light scattering profiles for anthropogenic sulphate aerosol alone will still require in situ aircraft monitoring to resolve. On average, the majority of clear weather data showed little, if any, aerosol structure to 2.4 km. The few cases of strong structure have allowed for the identification of three possible mechanisms which may control vertical profiles of haze.

a. Modification of aerosol structure following isentropic surfaces

Synoptic-scale warm air advection from midlatitudes modifies the inversion structure in the Arctic. During the March study, a case of warm air advection was studied in detail which showed the subsidence of a layer of aerosol as the inversion intensified. (A discussion of this event can also be found in a subsequent paper.) In Figure 4, the time–height display of the light scattering from the 6 March 1985 event is shown. The lower layer of aerosol during the morning of 6 March was vertically compressed by the lowering of the surface inversion by the intrusion of warm (−14°C) air aloft. Figure 5 shows a time–height diagram of temperature for the March study period and shows the warm air intrusion from 500 m to 3 km at 0000 UTC 7 March. The gradient in lidar backscattering at the top of the layer followed a 260 K potential temperature isotherm. At the end of the period of warm air aloft, clearing was accompanied by strong cooling from 500 m to 3 km. Over the next 24 h the inversion structure broke down giving a pulse of material at the surface in high winds. This is clearly a mechanism for transport of material aloft to the surface.

The overriding warm air could easily be interpreted as a warm front. Synoptic charts for the period do not show a strong frontal structure or even a system, but synoptic analysis in the Arctic is sketchy at the best of times because of poor meteorological data density. The

![Graph showing weekly average sulphate concentration measured at the Alert Station high-volume sampler. The lidar sampling weeks are marked by arrows.](image-url)
lack of aerosol in the warm air may indicate a different source region than the underlying colder (and probably older) more polluted air mass. What is apparent is that the intrusion of warm air lowered the maximum of the aerosol scattering to an altitude where surface effects and topography could play a more important factor in mixing the aerosol to the surface.

It has been a conjecture for several years that midlatitudinal aerosol transport to the Arctic may be governed by isentropic surfaces rather than assuming isobaric transport (Carlson, 1981; Iversen, 1984). This observation and the following section provide some evidence for that possibility, at least on the local scale.

b. Foehn (chinook)

Over the 5–7 February period, a layer appeared at an altitude of 3.6 km, descended to an altitude of 1.6 km and then rose to the original height over a period of 36 h (Fig. 6). Figure 7 shows time–height diagrams of temperature and humidity for the period over which the foehn occurred. Since these diagrams are smoothed somewhat, the actual temperature, relative humidity and water vapor pressure from the fifth to the seventh are shown in Figure 8. The layer was in extremely dry air (<60% relative humidity over water over most of the period and 28% relative humidity at its minimum at 1200 UTC on 6 February) and unsaturated with respect to ice. The air was extremely warm (−8°C at the center of the period while the surface was −35°C). Trajectories for the entire period (Fig. 9) indicate flow off the Greenland ice cap. The combination of features make it quite likely that this event is a foehn (chinook) from subsiding air off the ice cap. Synoptic analysis shows that a strong low existed on the southeast coast of Greenland and that a smaller low slid up into lower Baffin Bay and the Davis Strait during the end of the period. If the event were a system approaching from the southeast of Alert, it would be expected that it would contain more moisture (such as the March 1985 event) than was seen here. The lack of moisture also indicates subsidence from high in the troposphere.

![Rayleigh Ratio](image)

**FIG. 3.** Average weekly $R$ profiles for the November–February studies.

![Time–height temperature diagram](image)

**FIG. 5.** Time–height temperature (°C) diagram for the March 1985 study period. Time tick marks represent 0000 UTC.
Fig. 6. Time-height color display of the 5-7 February chinook event.
Fig. 7. Temperature (°C) and humidity (%) time-height contours for the first week of February, 1985. Date marks represent 0000 UTC. The warm, dry air intrusion represents the chinook.

Fig. 8. Temperature, relative and absolute humidity soundings for the 5–7 February period: 5 February, 0000 and 1200 UTC, circles and triangles, respectively; 6 February, 0000 and 1200 UTC, diamonds and pluses, respectively; and 7 February, 0000 and 1200 UTC, x’s and stars, respectively.
Fig. 9c. As for 9a, but arriving at 1200 UTC 7 February. The final 6 h of flow at the three levels show a southwesterly component and the 850 mb flow does not come from Greenland.
The general trajectory and flow across Greenland were undoubtedly influenced by the North Atlantic low.

The lidar data shows that the subsidence has provided a mechanism for vertical transport of aerosol to near the surface. Measurements of $D$ over this period indicated that the layer consisted of spherical scatterers. Given the relative humidity, this rules out ice crystals or water droplets and indicates that the layer is probably dry particulate. This event, however, remained completely decoupled from the surface, and the SO$_4^{2-}$ concentrations at the high-volume air sampler (205 m MSL) were low during this week. By the seventh, the 850 mb trajectories begin to show a southwesterly influence which would have less downslope flow, and cooling occurred with the weakening of the inversion over the lowest 4 km. The water vapor density increased by 0.1–0.2 g m$^{-3}$ from 1200 UTC 6 February to 1200 UTC 7 February and this, coupled with the cooling, caused a large increase in the relative humidity (to ice supersaturation) above 1500 m after 0000 UTC on seventh. Snow began at 3800 m where the air was water saturated. Snow descended to the surface by 1100 UTC.

(Ths, the strong scattering on the right side of the Fig. 6.)

The identification of this event as a foehn may be challenged given the sparse data in the Arctic and the lack of a system on the synoptic charts. Clearly, however, a dry, warm mechanism is operating here which shows the potential of moving aerosol vertically throughout much of the troposphere over a 3 day period. Should the 1600 m minimum of the aerosol be eroded by inversion breakdown or mechanical mixing from orography, material originally at a height of 3 to 4 km could find its way to the surface.

4. Ice crystal light scattering

When it was clear from the magnitude of $R$ and $D$ that submicrometer aerosol scattering was being dominated by larger scatterers, it was often found that ice crystals were precipitating outside. In one striking case during the December dark period, it was possible to visually observe the lidar return from a cloud of crystals about 100 m above the ground. The return visually appeared as a multitude of “fireflies”, which, in reality, were the specular reflections from the plane surfaces of the crystals. In the March study when ice crystal replicas were photographed, it was found that the predominant form of the crystals were small columns and clusters of columns (rosettes), and small hexagonal plates.

On 19 November 1984, the temperature profile showed only a weak inversion from $-30^\circ$C at 100 m to $-28^\circ$C at 1600 m (Fig. 10). The air was highly saturated with respect to ice and nearly water saturated. At 0300 UTC on 19 November, a layer of ice crystals began to fall from above 2500 m and no cloud was noted by the observers. The 0000 UTC rawinsonde observation shows a $-5.1^\circ$C km$^{-1}$ lapse rate between 2300 and 3600 m, which is conditionally unstable. Over the next 2.5 h, five distinct showers of crystals fell (Fig. 11). The fall speed of the crystals was 0.4 m s$^{-1}$ indicating that they were large (>700 µm). Although the lidar profiles did not follow the crystals below 300 m, it is apparent from observer reports that the crystals did reach the surface.

It is of interest whether crystal showers such as these could scavenge aerosol from higher altitudes and leave them at lower altitudes. The layer seen at 800 m on 20 November (Fig. 1) lies just above the relatively dry air seen in Fig. 10 (+, × on the figure). The possibility exists that this layer was the nucleating core of the falling ice crystals which sublimed in this drier layer at a different location and time. The dry layer was not coincident with the showers at Alert. The crystals would have taken 8 min to traverse this dry layer and some could have sublimed in this time. This mechanism has been discussed by Hogan et al. (1984).

An alternative explanation could be that the layer was there all along and that the ice crystal showers merely obscured the layer from the lidar. The observations of Borys (1984) and those discussed by Barrie (1985) would suggest that ice crystal precipitation scavenging is not an effective removal mechanism for anthropogenic aerosols. The observations here (which are by no means conclusive) may encourage a deeper look at the problem of precipitation scavenging in the Arctic.

A confusing but common occurrence was the observation of depolarization ratios approaching and exceeding unity. Takano and Jayaweera (1985) have calculated these high depolarizations for randomly oriented hexagonal plates when the lidar is pointed about 30° from the vertical (cf. Fig. 9 of their paper). In this study, however, such depolarizations were seen at altitudes of 1–2 km with a vertically pointing system. Since the actual crystal habit and orientation in these layers are unknown, it is possible that columnar aggregates may also give such depolarizations. The fact that the on-and-off polarization components of $D$ are not measured simultaneously could also cause the value to be greater than unity in nonstationary conditions.

The effect ice crystals may have on the scattering of light can be estimated by looking at the relative cross sections for haze versus large crystals. Ignoring nonsphericity and ray optics effects in the crystals, one may calculate that 200 scatterers cm$^{-3}$ of 0.2 µm diameter, with a scattering efficiency of 6, would give a scattering coefficient of the aerosol of $3.75 \times 10^{-4}$ m$^{-1}$ (a reasonable level for the Arctic). One 200 µm ice crystal per liter of air (also a reasonable concentration) would give, with a scattering efficiency of the Fraunhofer limit of 2, a scattering coefficient of $3.14 \times 10^{-4}$ m$^{-1}$. (A similar computation was done for integrating nephelometer measurements in a preceding paper, Leaitch et al., 1984.) Thus, it is not difficult to see why unambiguous
Fig. 10. Temperature, relative and absolute humidity soundings for the 19-21 November period: 19 November, 0000 and 1200 UTC, circles and triangles, respectively; 20 November, 0000 and 1200 UTC, diamonds and pluses; and 21 November, 0000 UTC, X's.
Fig. 11. Height and width of the ice crystal precipitation band as a function of time on 19 November 1984. The rate of fall is the slope of the curve and a reference of 0.5 m s$^{-1}$ is noted on the figure. The layers were approximately 50–100 m thick.

Lidar measurements of Arctic sulphate aerosols are possible only in ice unsaturated air. It also points out that ice crystal scattering is a significant factor in the reduction of visibility along with the haze aerosols.

Since most of the winter study was done in the dark period, accurate visibility observations were not possible. However, it was seen in the March study (after sunrise) that cases of extremely reduced visibility were often accompanied by ice crystal scattering and precipitation. Using scattering parameters during periods of high $R$ which are more indicative of cloud than Mie scattering aerosols (Volkovskii et al., 1980; Barber and Larsen, 1985), it was found that excellent agreement between observed visibility and lidar measurements was possible (Fig. 12).

A final observation to be made after the winter study concerns the occurrence of cloud in the Arctic. At meetings on Arctic meteorology and air chemistry, the lack of cloud in the Arctic winter has been mentioned as a controlling factor in the lack of scavenging of aerosols in the Arctic. From the limited experience gained in this study, it is apparent that cloud (both mid- and low-level) is not that uncommon in the Arctic winter. There is, given the temperature, a lot of water available and near the surface the atmosphere is nearly always ice saturated. It has been stated to the author that the Arctic troposphere is essentially the stratosphere. This is based on the infrequency of neutral and unstable lapse rates in the Arctic winter. Certainly, much of the cloud seen in winter is cirrus-like and the ice crystal composition of these clouds make ground-based cirrus an apt description. Often these clouds are obvious on the lidar but go unreported by the meteorological observers because they are subvisible. The author has noted that the slight blurring of stars in the Arctic night and a simultaneous loss of refractive scintillation may be the only visual indication of these tenuous surface-based clouds. It is believed that ice crystal clouds are often available in the Arctic but are ineffective in scavenging aerosols, thus allowing the long Arctic residence times for anthropogenic aerosols.

The author believes the occurrence of fewer ice crystals in the months of October to December than in spring is caused by the lack of nuclei for their formation. From our experience in the spring months, ice crystals are prolific near the surface and this is probably correlated with increased haze and increased nuclei. Sulfate, the chemical species of greatest abundance in anthropogenic Arctic haze is not thought to be a good ice nucleus (Borys, 1984). However, increased sulphate may also parallel increased nuclei from midlatitudes.

5. Conclusions

During the course of the winter lidar study at Alert, some evidence was obtained to confirm that when there was structure to the aerosol backscattering profile, it was at an altitude of 500–1500 m above the surface, a height which corresponds well to the inversion. Such strong structures are the exception rather than the rule and the general light backscattering profile is relatively uniform. This study has indicated mechanisms for vertical haze transport. The first is isentropic. The haze tends to follow surfaces of constant potential temperature. If such an observation can be followed back to
the sources of the pollution (with proper consideration of air mass cooling along the path), it would explain why haze aerosols might be unaffected by surface deposition since the surfaces would tend to ride up into the troposphere as they move north.

The second mechanism influencing vertical motions is orographic, and has been identified with a foehn (chinook) off the Greenland ice cap. Such dry air subsidence near Greenland could bring air from above the 700 mb level to near the surface inversion. A third mechanism involves ice crystal scavenging and sublimation in dry air near the inversion. The evidence for this mechanism remains sketchy but does indicate that further work on precipitation mechanisms is needed.

Optical study of the Arctic troposphere is much more a study of ice crystal physics than a climatology of sulphate aerosol. It is clear that the predominance of the light scattering in the Arctic, especially low visibility cases, are due to ice crystal interference. Anthropogenic haze may be a contributor to those cases, but the maximum Rayleigh ratio due to anthropogenic aerosol alone was 7.

Acknowledgments. The author would like to thank Frank Froude for sharing the task of making nine separate trips to Alert for the purpose of this study. Keeping a lidar running at a location some 3000 km away from spare parts can be a challenging experience. The author would also like to thank Dr. Neil Trivett of the AES WMO BAPMoN facility who provided logistical support and the crew at the Alert Rawinsonde Station (H. Ewen, W. Davidson, and G. Stansfield) who were only too willing to help in interpreting the on-line lidar data. The author appreciates the permission by Dr. Len Barrie for use of the Alert sulphate data from 1984/85 and to Drs. Trivett, Barrie, J. P. Blanchet, R. E. Mickel, and G. DenHartog for the referral of some data which will also appear in a paper summarizing the March 1985 project. The support of the Canadian Armed Forces in providing transport and accommodation at Alert is gratefully acknowledged. The author would like to thank Dr. Brian Kerman for a review of the manuscript.

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