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A COMPARISON OF GROUND-LEVEL AND AIRCRAFT SAMPLING OF AEROSOLS AT BARROW, ALASKA

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ABSTRACT

We compare the analysis of Arctic haze aerosols collected at the NOAA-GMCC observatory near Barrow, Alaska, with results obtained by aircraft sampling during the "AGASP 1983" experiment. During this program, we collected many filter samples and operated the LBL aethalometer, an instrument that measures the aerosol graphitic ("black") carbon concentration in real time. From these results we obtain vertical distributions of various aerosol components that are in good agreement with the 24-hr averages measured at the ground station. We find substantial concentrations of black carbon and sulfur at all altitudes in the Arctic troposphere. The black carbon can be found in layers with peak concentrations significantly greater than the ground-level measurements.

INTRODUCTION

The Arctic haze has been studied in detail at various ground stations around the western perimeter of the Arctic. It appears to have a large combustion-derived component, containing particulate carbon in both organic and microcrystalline graphitic forms. (Rosen et al., 1981; Rosen and Novakov, 1983a). The latter species is often termed "black carbon." It is released into the atmosphere only as a by-product of

incomplete combustion and may have a long residence time. It is inert to atmospheric transformation, and so its presence in polluted atmospheres is a tracer for combustion emissions, usually of anthropogenic origin. In the Arctic, the optical absorption of this material appears to be large enough to cause a significant perturbation of the solar radiation balance, the magnitude of which will depend on the vertical and horizontal distributions of the aerosol (Porch and MacCracken, 1982; Cess, 1983). The haze also includes trace metals and sulfur species (Rahn and McCaffrey, 1979; Barrie et al, 1981; Ottar, 1981). To study the distributions and to link airborne measurements with the considerable data base obtained from ground stations, the AGASP 1983 flight patterns involved vertical and horizontal soundings upwind of Barrow, Alaska. Aerosol filter samples are routinely collected there at the NOAA/GMCC observatory, and this activity was intensified during the AGASP program so that 24-hr samples were taken for each day of the overflights (Bodhaine et al., 1984).

On board the aircraft, we collected many filter samples for periods of typically 1-2 hr, covering both level flight tracks and vertical profiles. We also operated the aethalometer, an instrument recently developed at LBL that responds in real time to the concentration of black carbon (Hansen et al., 1982). Analysis of the data enables (1) a comparison of the concentrations of aerosol black carbon and major and minor elements between the ground station and the atmosphere above it and (2) a determination of the distributions of these components. The results show that there is good agreement between the ground and aircraft measurements, that the aerosol extends horizontally to all locations sampled, and that the vertical distributions can show substantial

concentrations up to 10 km, the highest altitude attained.

EXPERIMENTAL DETAILS

Accompanying articles in this issue outline details of the AGASP 1983 program and of some of the other techniques, measurements, and results obtained. The LBL installation was designed to study the aerosol particle component of the Arctic haze and consisted of the aethalometer and two filter sampling lines.

The aethalometer collects the aerosol on a filter (Pallflex quartz fiber, type 2500 QAO) and uses an optical technique (Rosen and Novakov, 1983b) to measure the concentration of black carbon in real time. The filter in the aethalometer is changed from time to time to yield an integrated sample available for chemical analysis for black carbon. The sensitivity of the aethalometer is inversely proportional to its time resolution period: as used during AGASP, it had a detection limit of approximately 75 ng/m^3 of black carbon for a 1-min acquisition time. Vertical distributions are determined by grouping the data taken during slow descents into ranges of altitude. We then calculate the average black carbon concentration measured during the time spent within each range. The measurement accuracy is proportional to the total data acquisition time in the range, so the range boundaries are chosen for each flight profile to optimize the balance between accuracy and vertical resolution. Methods for the chemical and optical determination of the black carbon content of aerosol samples on filters have been developed at LBL (Gundel et al., 1984), from which we derive a calibration curve for the aethalometer.

In addition to the aethalometer, we also operated two filter sampling lines with which we collected a large number of total aerosol samples on cellulose filters (Millipore, type RATF) and prefired quartz fiber filters (Pallflex, type QAO 2500). These were operated at airstream face velocities of 1.5 m/sec and 5 m/sec respectively, enabling the collection of sufficient material for analysis in periods of 1-2 hr. These analyses include the determination of black carbon and the categorization of organics by combined optical and thermal evolved gas analysis, and the measurement of major and minor elements by x-ray fluorescence. Together with the quartz fiber filters removed periodically from the aethalometer, a total of 47 filter samples were taken during the four AGASP flights in the vicinity of Barrow, Alaska. By appropriately timing the filter changes relative to the flight plan, we collected samples whose air flows were primarily taken from particular altitude ranges. In the method we use here to present these data, the extent of each solid line parallel to the altitude axis indicates the range encompassing 70% of the total air volume sampled by a particular filter, with adjacent broken lines accounting for the next 20% of the air volume.

The aerosol samples collected at the NOAA/GMCC ground station were on filters run for 24-hr periods at airstream face velocities of approximately 1 m/sec, and were analyzed using the techniques mentioned above. Comparison of the results for the four AGASP overflight days with those for adjacent days shows no unusual features or concentration anomalies, suggesting that those four days yielded representative samples of the Arctic haze.

It should be noted that concentrations are reported here in units of nanograms per geometric cubic meter, rather than as mixing ratios, in order to interface directly with calculations of optical properties.

RESULTS

Table 1 shows some of the results from the analyses of the 24-hr ground level samples taken at Barrow on the AGASP flight days. Figure 1 shows the vertical distribution of black carbon obtained near Barrow during AGASP flight number 1. It is clear that there is a very pronounced layer below 1 km altitude, with peak concentrations between 400-500 m altitude. Note, however, that substantial concentrations of the order of 100 ng/m^3 are present up to 5 km altitude. In Fig. 2 we show the distribution obtained during flight 3: in this distribution, we detected a lower layer extending from ground level to 800 m altitude, then a region of relatively clean air, and then a broad upper layer having a maximum at approximately 1.5 km altitude and extending to 5 km. Although the peak concentration was less than one-half of that measured on flight 1, the total air column burdens were similar: 130 (±10) ng/cm^2 for flight 1, 0-10 km, and 135 (±15) ng/cm^2 for flight 3, 0-9 km. In both profiles, the vertical distributions show a reduction in concentration at the lowest levels relative to the peaks observed at altitudes of a few hundred meters, and the 24-hr average concentrations observed at the ground station were lower still. To reproduce the air column burdens derived from the aircraft soundings, it would be necessary to multiply the ground-level average concentrations by column lengths of 5.4 km and 6.4 km respectively.

In Figs. 3 and 4 we show results from the XRF analysis of the Millipore filter samples collected on flights 1-4. The concentrations of sulfur and bromine are plotted versus the altitude ranges encompassing the majority of each filter's air flow (solid lines, first 70%; broken lines, next 20%) and are compared to the 24-hr average values measured on the ground. The sulfur data presented in Fig. 3 show fairly substantial concentrations at high altitudes (of the order of 200-300 ng/m^3), increasing considerably at lower levels with the suggestion of a peak somewhat above the ground. The average ground-level concentrations are consistent with this profile. The peak concentration measured during flight 1 on the filter operated mainly at 1 km altitude is about 50% higher than the corresponding average ground measurement that day. Figure 4 shows the bromine data in the same format; due to lower concentrations and measurement interferences, not all filters yielded significant analyses and so less data are represented. The highest bromine concentrations on the ground and aloft were measured on March 11, 1983, the day of flight 1. However, in general the data show no discernible vertical structure except for a substantial reduction in concentrations at altitude relative to the ground-level measurements.

DISCUSSION

Perhaps the most striking features of these data are the intensity and vertical extent of the aerosol components. During AGASP flight 1, the peak black carbon concentration detected at 500 m altitude was more than five times greater than the average ground-level value and was in fact greater than the annual average concentration measured at LBL, located near the center of the urban San Francisco Bay Area. An illus-

tration of the vertical extent of the haze is provided by the fact that when expressed as a mass mixing ratio, the black carbon concentration measured during AGASP flight 3 between 4 km and 5 km altitude was 50% greater than the average ground-level measurement that day. Air masses at different altitudes may have passed over different source regions, and the presence of substantial quantities of black carbon at 5 km altitude may suggest a quite distant origin. The fact that we measured greater concentrations of black carbon at moderate altitudes than at ground level suggests that the concentrations of this material in the Arctic haze may indeed be generally larger than is indicated by the data base of ground-level measurements.

The sulfur data shown in Fig. 3 present some evidence of a peak in concentration at low altitudes but also show that we measured substantial concentrations of filterable aerosol sulfur throughout the Arctic troposphere to 10 km altitude. The downward diffusion of sulfuric acid droplets present in the stratosphere as a result of recent volcanic activity (Gandrud et al., 1983) could be one contributor to this latter observation. The ground-level concentrations of aerosol sulfur were comparable to those measured in the lower troposphere, but the filter sequencing technique provides insufficient resolution for a more detailed study of the vertical distribution.

The results shown in Fig. 4 for the concentrations of filterable aerosol bromine show no discernible vertical structure but do show that the ground-level concentrations were always substantially larger than those measured aloft. These observations would be consistent with a ground-level natural source. (Berg et al., 1983)

CONCLUSIONS

During the AGASP 1983 flight program, we obtained real-time measurements of black carbon concentrations from which we derive vertical distributions of this component of the Arctic haze. Results obtained near Barrow, Alaska, showed surprising intensity and extent and indicated stratification of the polluted air masses. Black carbon has a large optical absorption coefficient, and the total air column burdens observed may cause a significant perturbation of the radiation balance (Rosen and Hansen, 1984).

We also collected aerosol samples on filters and analyzed some of these taken at different altitudes for sulfur and bromine. The sulfur data show large concentrations at high altitudes that may indicate a high-level source. The bromine data show less detail but are consistent with the hypothesis of an area-wide ground-level source.

Comparison of the aircraft results to those obtained from the NOAA/GMCC ground station at Barrow shows good agreement, indicating the validity of this site as a suitable location for the ground-level sampling of the Arctic haze.

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FIGURE CAPTIONS

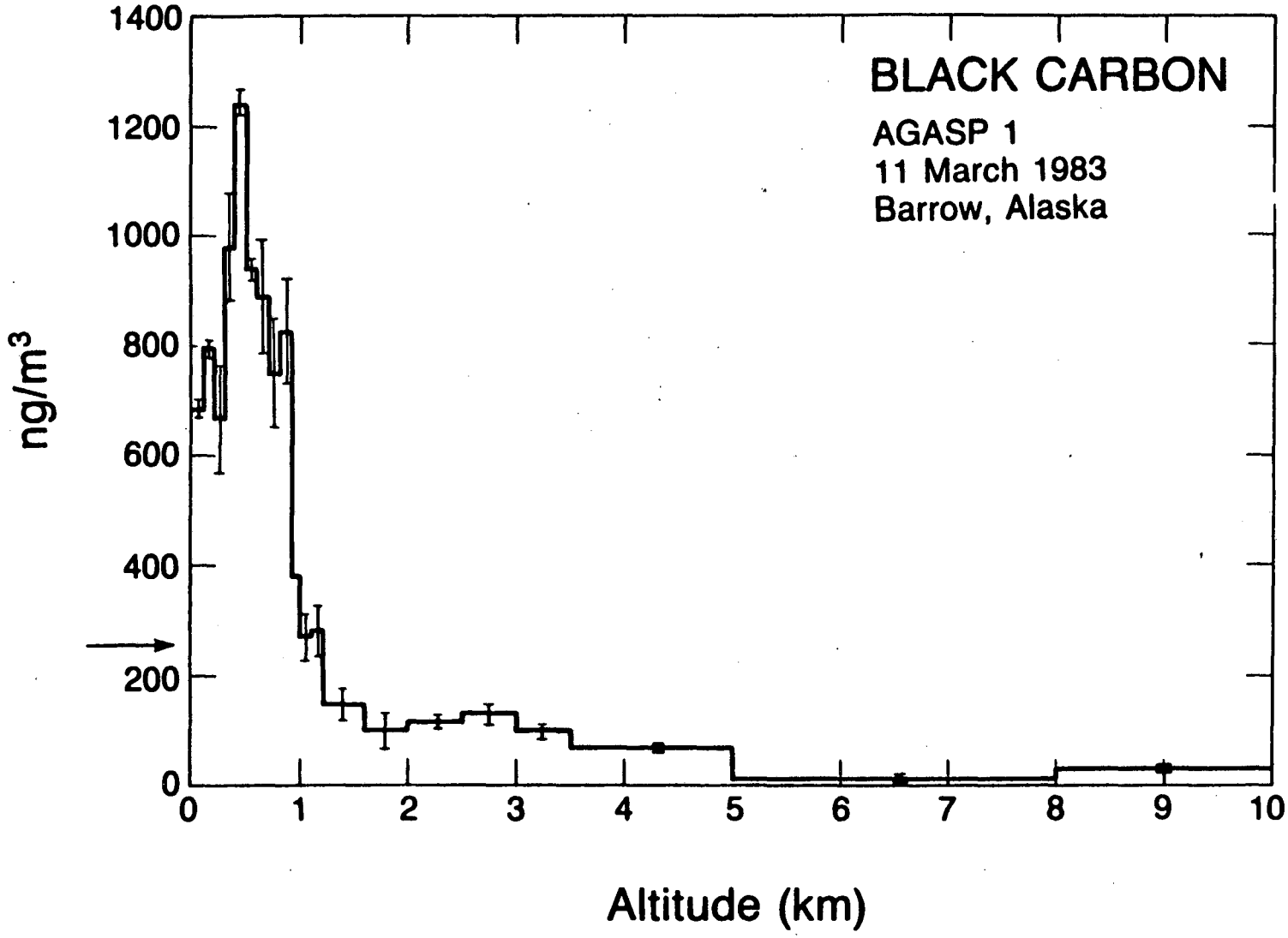
Figure 1. Vertical profile of black carbon concentration obtained during AGASP flight 1 on 11 March 1983 near Barrow, Alaska. The arrow on the abscissa indicates the 24-hr average black carbon concentration measured that day at the NOAA/GMCC ground station at Barrow. Units are nanograms per geometric cubic meter.

Figure 2. Vertical profile of black carbon concentration obtained during AGASP flight 3, 15 March 1983, presented in the same format as Figure 1.

Figure 3. Results from analysis of sulfur on Millipore filter samples collected during AGASP flights 1 to 4 near Barrow, Alaska, from 11 to 17 March 1983. Solid lines parallel to the altitude axis represent the range within which the first 70% of the volumetric air flow was sampled; broken lines encompass the next 20%. The error bar is placed at the center of the range covering the majority air flow, and the adjacent numeral indicates the AGASP flight number. The numbered arrows on the abscissa indicate the 24-hr average sulfur concentrations measured at the NOAA/GMCC ground station on the days of those flights. Units are nanograms per geometric cubic meter.

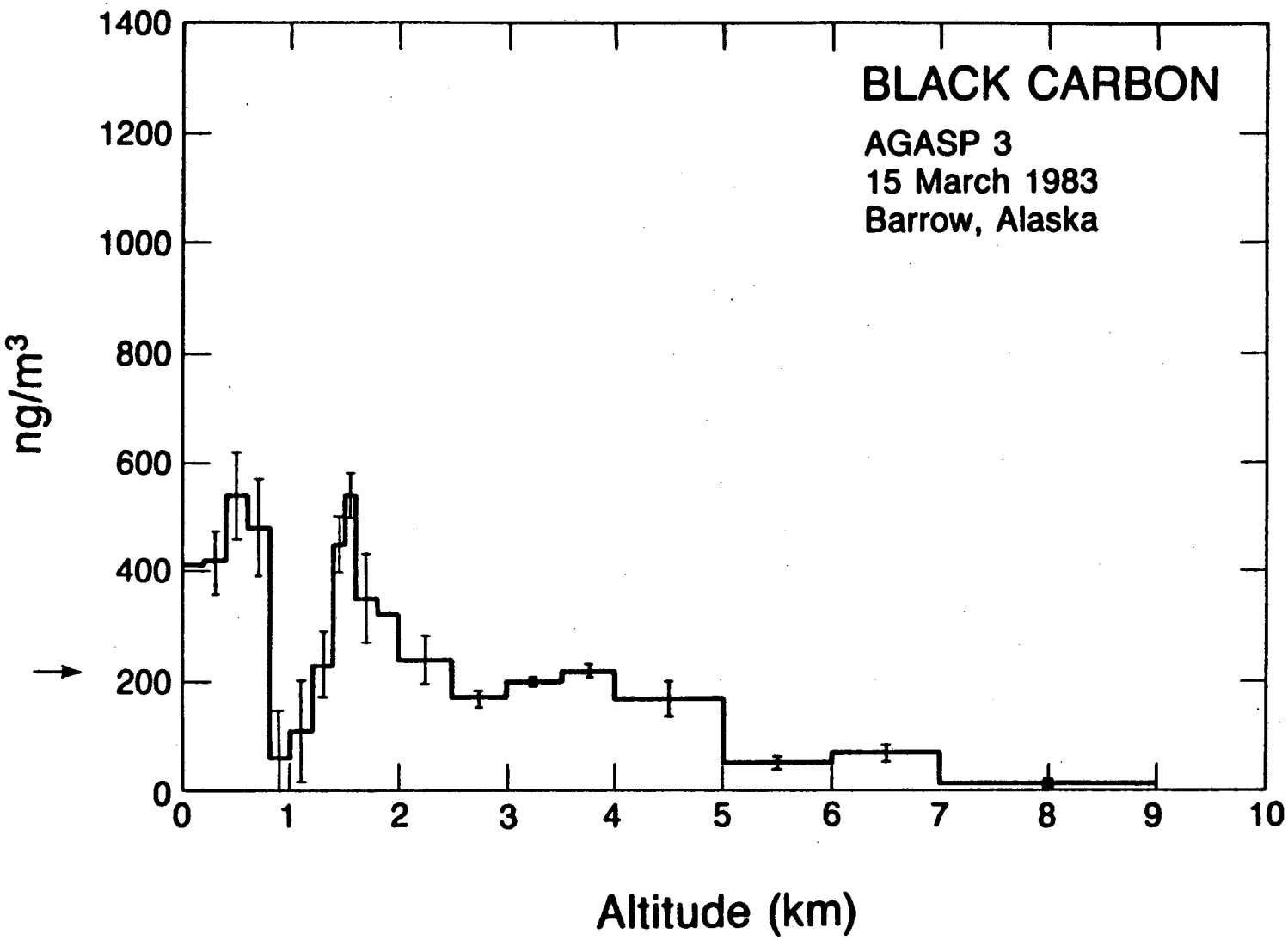
Figure 4. Data from the analysis for bromine on Millipore filter samples collected during AGASP flights 1 to 4, presented in the same format as Figure 3.

Table 1. Analysis of 24-hour aerosol samples collected at Barrow.



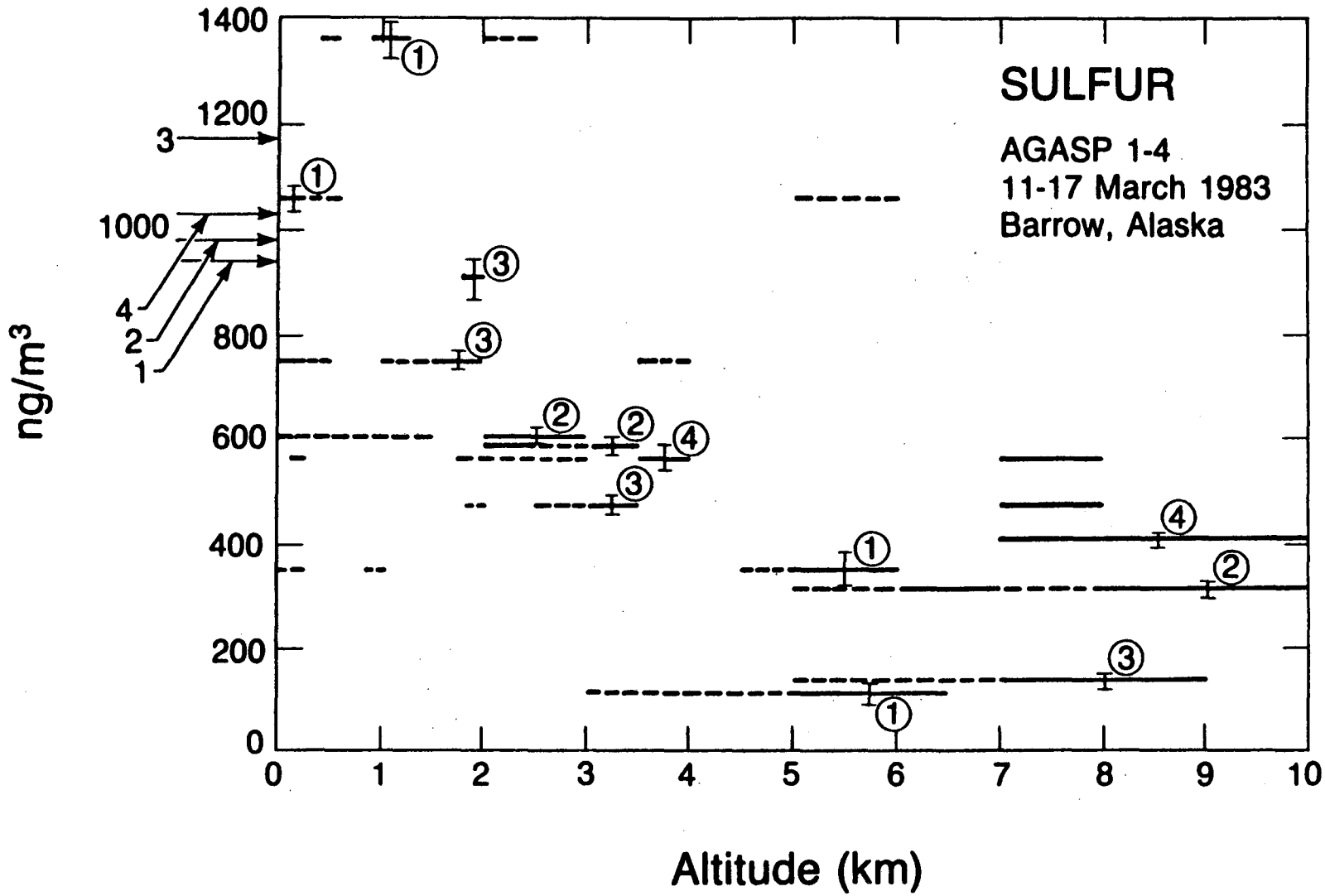
XBL 8312-6876

Figure 1



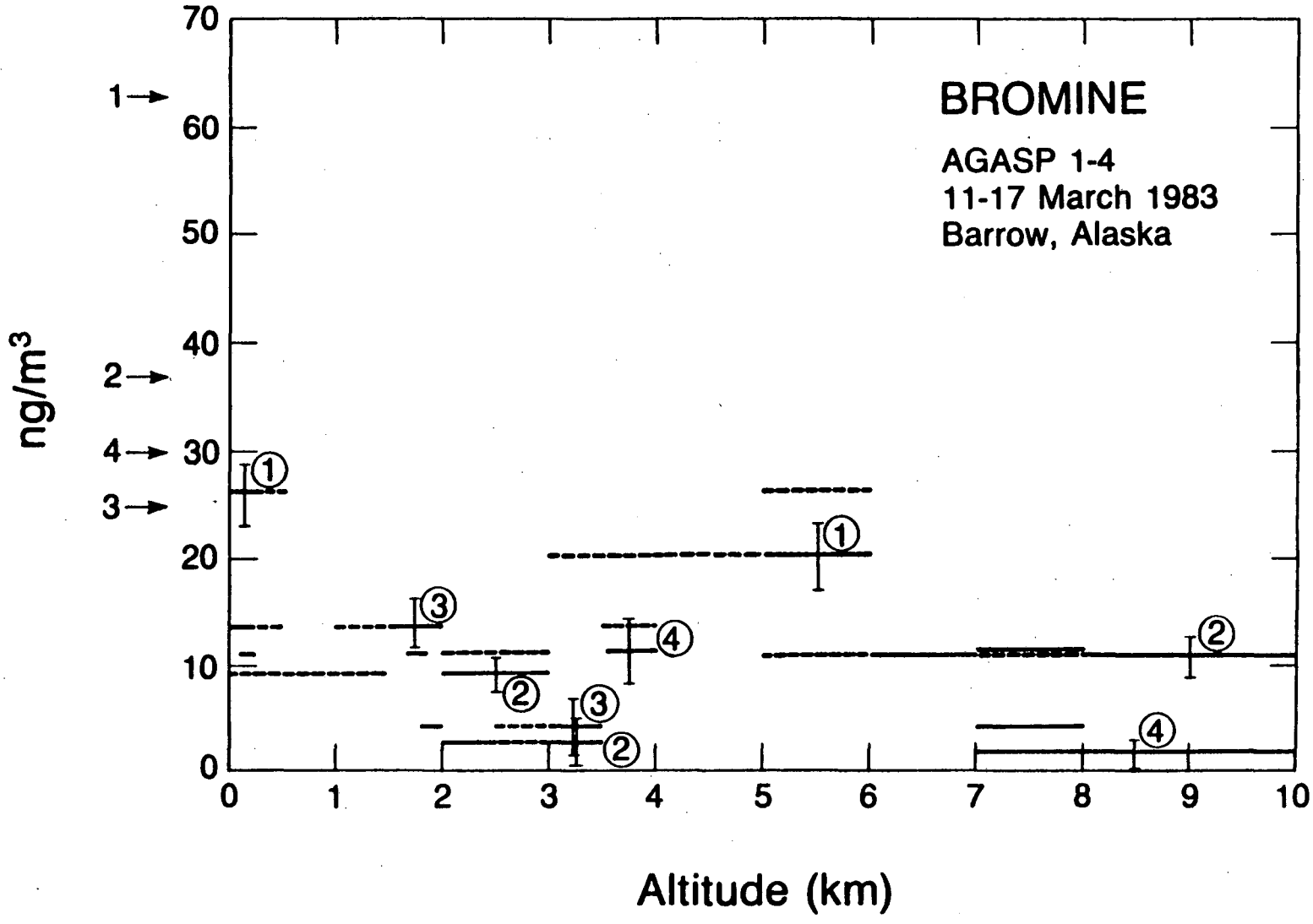
XBL 8312-6877

Figure 2



XBL 8312-6875

Figure 3



XBL 8312-6878

Figure 4

Table 1. Analysis of 24-hour aerosol samples collected at Barrow.

AGASP flight no.	Date	Concentration, ng/m^3				
		Black carbon	S	Br	Si	K
1	11 March 1983	241	940	63	65	41
2	13 March 1983	295	980	37	58	29
3	15 March 1983	206	1170	25	64	22
4	17 March 1983	253	1030	30	68	29
	Mean	249	1030	39	64	30

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