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VERTICAL DISTRIBUTIONS OF PARTICULATE CARBON, SULFUR, AND BROMINE IN THE ARCTIC HAZE AND COMPARISON WITH GROUND-LEVEL MEASUREMENTS AT BARROW, ALASKA

A.D.A. Hansen and H. Rosen

Applied Science Division, Lawrence Berkeley Laboratory, University of California

Abstract. We present results obtained during the "AGASP 1983" Arctic haze aircraft sampling experiment. We collected many filter samples and operated the LBL aethalometer, an instrument that responds to the aerosol graphitic ("black") carbon concentration in real time. We found substantial concentrations of black carbon and sulfur at all altitudes in the Arctic troposphere. Maxima in the black carbon concentrations were observed at intermediate altitudes. Comparing these results with data from the NOAA-GMCC observatory near Barrow, Alaska, we find that peak concentrations aloft can be significantly greater than the ground-level measurements.

Introduction

The Arctic haze 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"; its only source is incomplete combustion, and its presence in polluted atmospheres is a tracer for combustion emissions, usually of anthropogenic origin. In the Arctic, the optical absorption of this material could be large enough to cause a significant perturbation of the solar radiation balance [Porch and MacCracken, 1982; Cess, 1983; Rosen and Hansen, 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 included 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-h samples were taken for each day of the overflights [Bodhaine et al., 1984].

Experimental Details

On board the aircraft, we operated the aethalometer, an instrument recently developed at LBL that responds in real time to the concentration of black carbon [Hansen et al., 1982]. It 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. The filter is changed from time to time to yield a sample available for chemical analysis. Methods for the determination of black carbon in aerosol samples have been developed at LBL [Gundel et al., 1984], giving a calibration curve for the aethalometer. Vertical distributions of black carbon concentration are determined by grouping the data taken during slow descents into ranges of altitude and calculating the average. 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.

We also used two filter sampling lines to collect a large number of total aerosol samples on cellulose filters (Millipore, type RATF) and prefired quartz fiber filters (Pallflex, type QAO 2500). Analyses included 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. 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 24-h aerosol samples collected at the NOAA/GMCC ground station 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, for use in calculations of optical properties, concentrations are reported here in units of nanograms per geometric cubic meter, rather than as mixing ratios.

Results

Figure 1 shows the vertical distribution of black carbon obtained near Barrow during AGASP flight 1. There is a very pronounced layer below 1 km altitude, with peak concentrations between 400-500 m altitude; substantial concentrations of the order of 100 ng/m^3 are present up to 5 km altitude. Figure 2 shows data obtained during flight 3. 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 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. Flights 2 and 4 showed less pronounced vertical structure, but comparable extent and intensity of black carbon concentration. In all profiles, the vertical distributions show a reduction in concentration at the lowest levels relative to the

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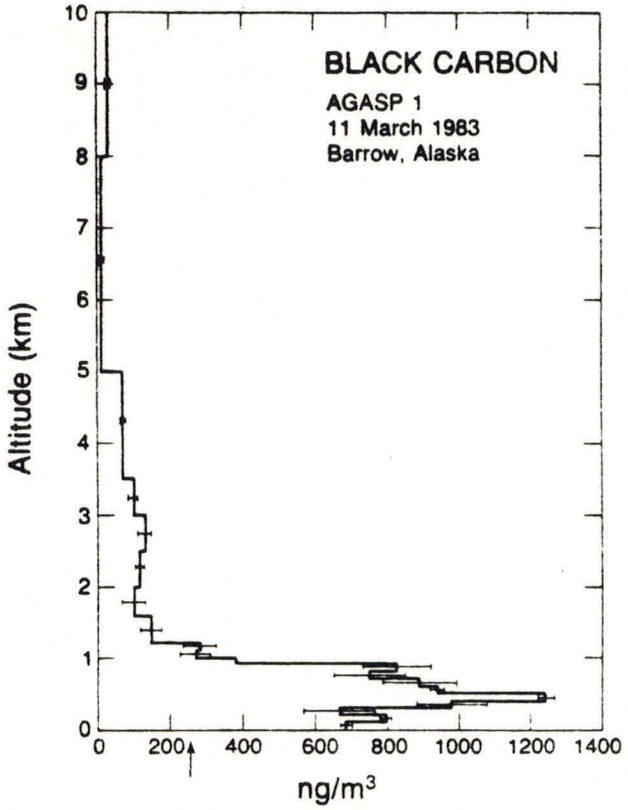

Fig. 1. Vertical profile of black carbon concentration obtained during AGASP flight 1 on March 11, 1983, near Barrow, Alaska. The arrow on the ordinate indicates the 24-h average black carbon concentration measured that day at the NOAA/GMCC ground station at Barrow. Units are nanograms per geometric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

peaks observed at altitudes of a few hundred meters, and the 24-h average concentrations of black carbon observed at the ground station were lower still. Table 1 shows some of the results from the analyses of the 24-h ground-level samples taken at Barrow on the AGASP flight days. To reproduce the air column burdens derived from the aircraft soundings on flights 1 and 3, it would be necessary to multiply those days' ground-level concentrations by column lengths of 5.4 and 6.4 km.

Figures 3 and 4 show results from the sulfur and bromine analysis of the Millipore filter samples collected on flights 1-4 compared with the 24-h average values measured on the ground. The sulfur data show concentrations of the order of 200-300 ng/m^3 at high altitudes, increasing considerably at lower levels with the suggestion of a peak somewhat above the ground. 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; not all filters yielded significant analyses, so less data are represented. The highest bromine concentrations on the ground and aloft were also measured on the day of flight 1.

Discussion

The intensity and vertical extent of the aerosol components are remarkable. During AGASP flight 1 near Barrow, Alaska, the peak black carbon concentration detected at 500 m altitude was more than five times greater than that day's ground-level average and was greater than the annual average concentration measured at Lawrence Berkeley Laboratory. When expressed as a mass mixing ratio, the black carbon concentration measured during 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 these altitudes could suggest a distant origin.

The sulfur data shown in Figure 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.

The results shown in Figure 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].

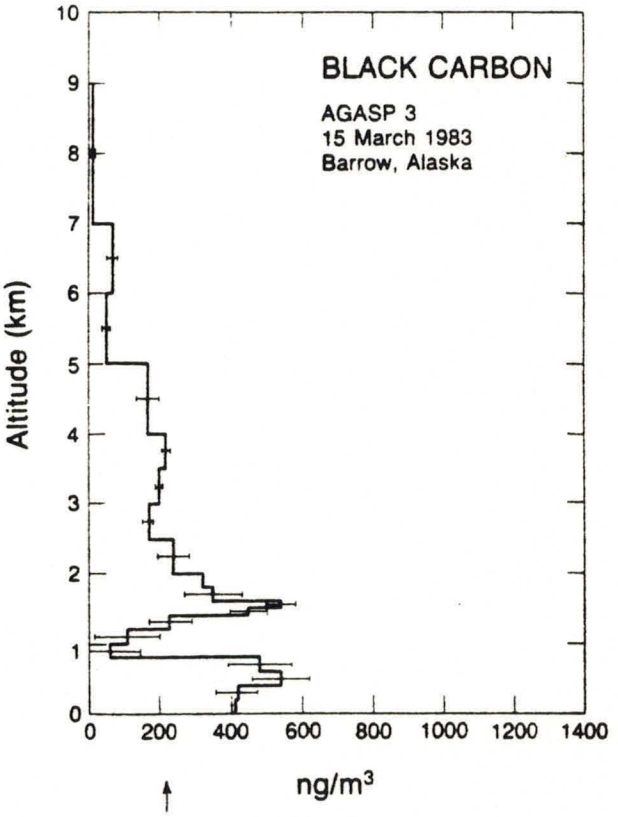

Fig. 2. Vertical profile of black carbon concentration obtained during AGASP flight 3, March 15, 1983, presented in the same format as Fig. 1.

TABLE 1. Analysis of 24-h aerosol samples collected at Barrow.

AGASP flight	Concentration, ng/m^3				
	Black carbon	S	Br	Si	K
1	241	940	63	65	41
2	295	980	37	58	29
3	206	1170	25	64	22
4	253	1030	30	68	29
Mean	249	1030	39	64	30

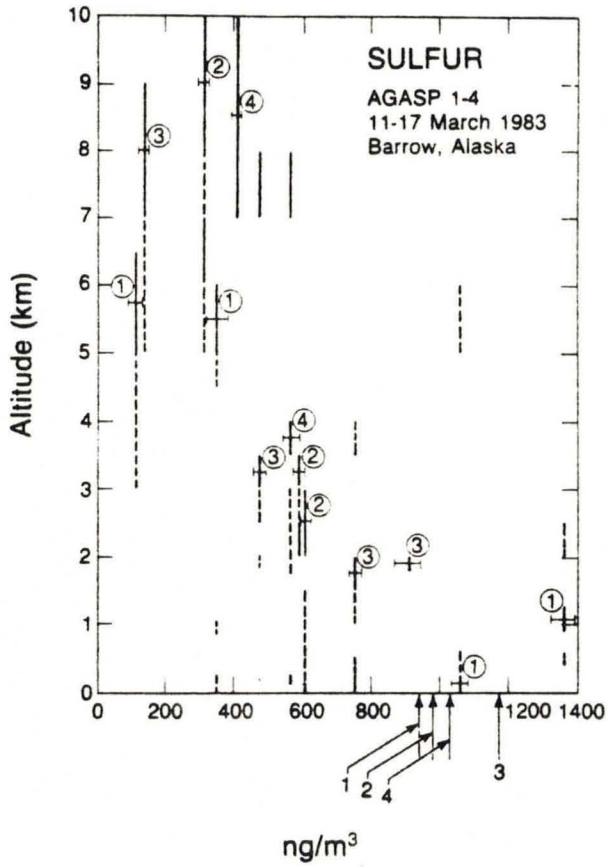

Fig. 3. Results from analysis of sulfur on Millipore filter samples collected during AGASP flights 1 to 4 near Barrow, Alaska, from March 11 to 17, 1983. Solid lines parallel to the altitude axis represent the range within which the first 70% of the volumetric air flow was sampled; broken lines encompass the next 20%. The error bar, representing 3 σ limits, 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 ordinate indicate the 24-h average sulfur concentrations measured at the NOAA/GMCC ground station on the days of those flights. Units are nanograms per geometric cubic meter.

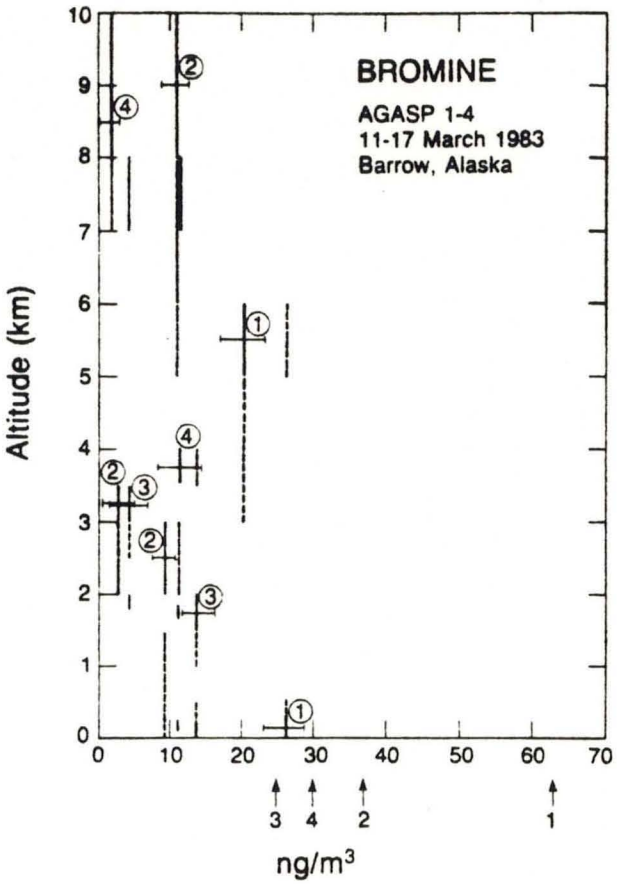

Fig. 4. Data from the analysis for bromine on Millipore filter samples collected during AGASP flights 1 to 4, presented in the same format as Fig. 3.

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, 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.

We also collected aerosol samples on filters and analyzed these 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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A.D.A. Hansen and H. Rosen, Applied Science Division, Lawrence Berkeley Laboratory, University of California, Berkeley, California 94720.

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