*Journal of Atmospheric Chemistry* **9**: 347–361, 1989.


# AEROSOL BLACK CARBON MEASUREMENTS IN THE ARCTIC HAZE DURING AGASP-II

A.D.A. Hansen and T. Novakov

Atmospheric Aerosol Research Group
Applied Science Division
Lawrence Berkeley Laboratory
University of California
Berkeley, California 94720

**Abstract.** During the second Arctic Gas and Aerosol Sampling Program conducted in April 1986, we performed measurements of the optically absorbing carbonaceous component of the ambient aerosol from the NOAA WP-3D aircraft operating between sea level and 10 km altitude. We collected the aerosol on filters that were exposed for several hours; we also operated the aethalometer to measure the concentration of aerosol black carbon in real time. The filter analyses represent averages over the altitude range and time span during which the filter was collecting. The real-time results were sorted by altitude to calculate vertical profiles of black carbon concentration. Values typically ranged from 300 to 500 ng $m^{-3}$ at lower altitudes, decreasing gradually to 25 to 100 ng $m^{-3}$ at 8-10 km. Strong stratification at lower altitudes was frequently observed. The magnitude of these concentrations suggests that the sources are distant regions of considerable fuel consumption. The presence of this material in the tropospheric column and its probable deposition to the high-albedo surface may result in perturbations of the solar radiation balance. The concentrations measured at the highest altitudes may mean that particulate carbon and accompanying emissions for which it is a tracer are mixing into the stratosphere.

Key words: Aerosol black carbon, Arctic atmosphere, Arctic haze, combustion, AGASP, optical depth.

## 1. Introduction

The main global source of energy production is the combustion of solid, liquid, or gaseous carbonaceous fuels (Marland et al., 1985). This results in the discharge to the atmosphere of the major species carbon dioxide and water vapor and of minor and trace effluents. These latter species include emissions of both gaseous and particulate pollutants that are strongly dependent on the nature of the fuel, the quality of the combustion, and the extent and efficiency of emission controls. One combustion byproduct is graphitic or "black" particulate carbon emitted as aerosol particles of submicron size. This material is a good tracer for combustion emissions: it cannot be

produced by secondary mechanisms from precursors in the atmosphere. It can have a long atmospheric lifetime, resulting in its transport to and detection at all parts of the globe (Andreae, 1983; Wolff, 1984); a recently started program is measuring aerosol black carbon on a daily basis at the South Pole Observatory (Hansen et al., 1989).

In winter months, the demands of space heating may result in greatly increased emissions in certain northern hemisphere regions. Favorable meteorological conditions may transport these emissions to all parts of the Arctic Basin. The Arctic haze has been documented for many years at numerous locations in the western hemisphere and appears to contain carbonaceous, silicaceous, and sulfate components, all of which could be related to combustion emissions (Rahn, 1981, 1985; Barrie, 1986). The fuel consumption within the region is insufficient to account for the observed concentrations of these species (Rosen and Hansen, 1985). Therefore, long-range transport from intermediate latitudes to the Arctic is indicated.

The presence of carbonaceous particles in the atmosphere may lead to a number of possible chemical or physical consequences. The carbon surface may contain a variety of adsorbed organic compounds and, in the presence of water, may act as a site for catalytic $SO_2$ oxidation (Brodzinsky et al., 1980). A possible mechanistic link between particulate carbon concentrations and the formation of aerosol sulfate has been deduced from the examination of wintertime data from a highly polluted urban location (Bizjak et al., 1986). In addition, aerosol black carbon has a very large cross section for the absorption of visible light, much larger than that of other aerosol species (Faxvog and Roessler, 1978; Rosen et al., 1978; Japar et al., 1986). This absorption may lead to optical depths sufficient to produce a perturbation in the solar radiation balance both in the general troposphere (Coakley et al., 1983), and especially over the highly reflecting winter Arctic surface (Shaw and Stamnes, 1980; Cess, 1983; Valero et al., 1984). Deposition of the black carbon to a snow or ice surface may affect the surface albedo and hence the thawing rates (Warren, 1984; Clarke and Noone, 1985). Finally, under certain conditions, carbonaceous aerosols may act as condensation nuclei, altering the size distribution and optical properties of clouds (Penner and Edwards, 1986; Pueschel et al., 1981; Twomey, 1977).

The AGASP-I program, conducted in 1983, demonstrated the presence of large stratified concentrations of pollutant species of anthropogenic origin in the Arctic haze (Schnell, 1984), including peak concentrations of aerosol black carbon that compared in magnitude with concentrations commonly measured in rural and suburban locations. In the second AGASP program, we fielded a more sensitive and more rapidly responding instrument in order to further study the distribution of this important component of combustion emissions.

## 2. Experimental Details

Our installation aboard the NOAA WP-3D aircraft consisted of the aethalometer and a bank of filters switched by solenoid valves; both components collected the ambient aerosol without size segregation. The aethalometer is an instrument developed at LBL that responds to the concentration of aerosol black carbon in real time (Hansen et al., 1982). The inlet airstream is drawn at a high flow rate through a prefired quartz fiber filter illuminated by a light source. The rate of decrease of light transmitted through the filter (relative to a reference beam) is proportional to the rate of accumulation of aerosol black carbon on the filter. With signal inputs for altitude and air flow rate (nominally 40 SLPM at sea level), the system calculates black carbon concentrations in real time and can print or display them as time series or accumulating vertical profiles. Periodically, the aethalometer filter is changed. The total accumulation of black carbon on the filter spot may be determined by laboratory analytical techniques (Groblicki et al., 1983; Gundel et al., 1984) and compared to the integral of the real-time output to confirm the calibration. The resu from this filter yields an average black carbon concentration for the altitude range and tir period covered. The switched filters were operated for several hours in different altitude rang to provide integrated samples for comparison with the real-time data. With the exception of t flight on April 14, 1986, good real-time data were collected for about 7 hours each flight. addition, two filter samples were collected each flight, and their analyses agreed well with t real-time data. The data are all reported in concentration units of nanograms per standard cub meter. The estimated errors on the measurements presented in this paper amount to a total of 25% and are due to the following uncertainties. An error of ± 10% is contributed by inaccuracy the air mass flow measurement. An error of ±5% is contributed from the calibration of the rel tionship between optical attenuation measurements and chemical analysis of filter deposits. A error of ± 10% is attached to the accuracy of our laboratory determination of carbon versus othe in an interlaboratory comparison (Groblicki et al., 1983). The first of these terms can lead to di tortion of vertical profiles or error in comparing different flights' results. The latter two ter lead to an error in the overall scaling of the data. In addition, groupings of data that represent total of only a few minutes of measurement are subject to instrumental noise; these results a shown in the profile figures with thinner (i.e., less emphasized) lines. The data presentatio scheme is discussed in the caption to Fig. 1.



## 3. Results

The study area, aircraft flight tracks, and profiles are discussed by other authors (e.g., Schnell al., this issue). The results of our measurements of aerosol black carbon follow.

*Flight 1, April 2, 1986: Alaskan North Slope*

Real-time data were collected for 359 minutes on this flight. The coverage was best below 400 m altitude, but considerable ferry time was spent at higher altitudes. Two filters were collecte one for most of the flight time below 6 km altitude, and the other between 1.2 and 1.8 km alt tude. The overall mean black carbon concentration was 310 ng $m^{-3}$; however, concentrations more than double this amount were observed at lower altitudes. Figure 1(a) shows the distributio of data recording time vs. altitude; Fig. 1(b) shows the deduced vertical profile of black carbo averaged over the entire flight. These data clearly show a large concentration of black carbo below 2.5 km altitude; a layer of cleaner air above that, about 1 km thick; and then increasin carbon concentrations up to about 5 km altitude. Unfortunately there was not enough coverage the altitude range in the 5-7 km range to follow the structure of the upper layer. The filter result are superimposed on the figure and show good agreement.

*Flight 2, April 8, 1986: Canadian North Slope*

During this flight, the system recorded data for 449 minutes. Good coverage of altitudes wa obtained at higher and lower levels, but only a few minutes of data were recorded between 3 kr and 6 km altitude. Figure 2(a) shows the vertical distribution of data recording time. The overa mean black carbon concentration was 200 ng $m^{-3}$. Two filters were collected - one below 6 kr altitude, and the other above it. Mean concentrations derived from the filter analyses seem some what low when compared to the real-time data. In Fig. 2(b) the deduced vertical profile of blac carbon concentration is shown, with the filter results superimposed. The overall vertical distribu tion shows generally higher concentrations at altitudes below 3 km but with black carbon bein detected at all levels.

*Flight 3, April 9, 1986: Alaskan North Slope*

We recorded 523 minutes of data on this flight, 3½ hours of which were at ferry altitudes and th

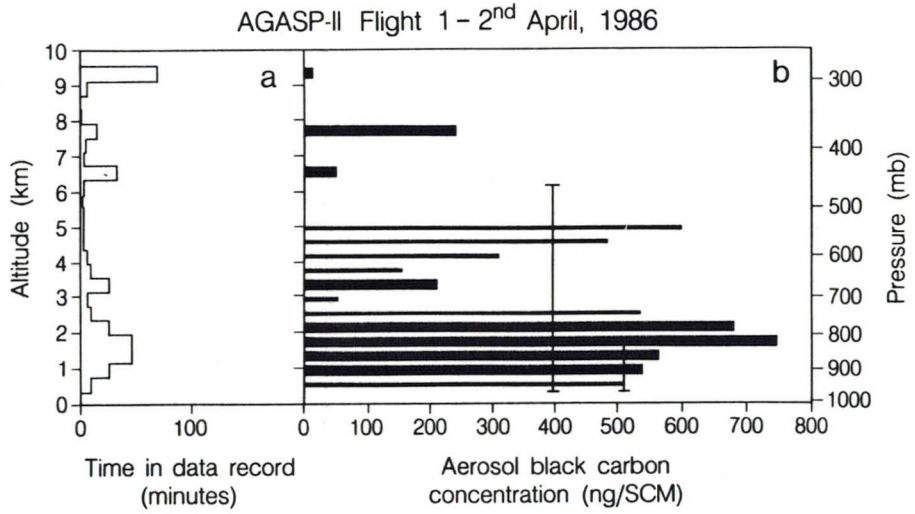


Figure 1. (a) Distribution of data recording time and (b) vertical profile of black carbon concentration averaged for the data set of AGASP-II flight 1. The vertical lines indicate the mean concentrations calculated from two filters' analyses, and extend over the altitude range in which 75% of the filters' air flow was collected. The horizontal lines represent the averages of the real-time data collected into altitude ranges of 400 m extent. Some altitude range bins for the profile data contained only a few minutes of measurement coverage, resulting in the inclusion of instrumental noise in the measurement uncertainty. These data are represented by thinner horizontal lines. With greater than about 20 minutes of data collection time per altitude bin, the measurement error is approximately ± 15%, dominated by calibration and airflow measurement uncertainties (e.g., data around 1 km, shown by heavy lines). With 8-20 minutes of data, instrumental noise increases the error to approximately ± 20 - 50% (e.g., data between 4 and 5 km, shown by medium lines). With less than 8 minutes of data in a bin, the instrumental noise can result in errors of ± 50-100%. If other factors render the data significant, they are represented by the thinnest lines (e.g., profile in Figure 4); if the errors are too large, the data are not shown. Units are nanograms per standard cubic meter.

remainder representing good coverage of lower altitudes and modest coverage in the 3-6 km range. Figure 3(a) shows the vertical distribution of data recording time. We collected two filter samples, above and below 6 km. The overall mean black carbon concentration was 230 ng $m^{-3}$. Figure 3(b) shows the vertical distribution of black carbon averaged over the whole flight. The data indicate concentrations of around 150 ng $m^{-3}$ above 6.5 km altitude, a cleaner layer at about 6 km, then a more polluted layer centered at about 4.5 km altitude. Below this level the concentrations diminish to a minimum in the 1200-1600 m range but then increase sharply in a low-lying layer, which in turn does not appear to extend to ground level. Thus we see that the data suggest a stratification of the atmosphere into at least three polluted layers separated by two regions of cleaner air.



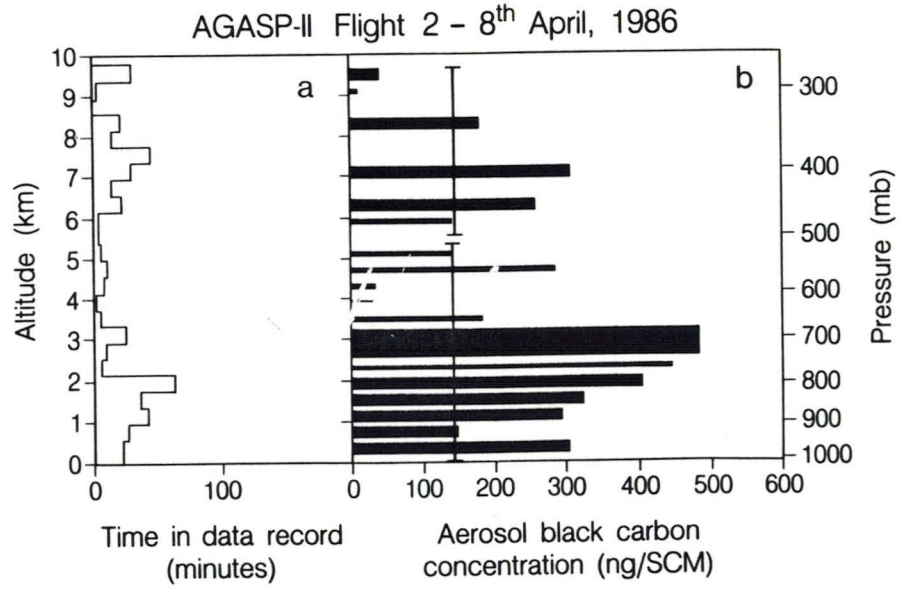


Figure 2. (a) Distribution of data recording time and (b) vertical profile of black carbon concentration averaged for the data set of AGASP-II flight 2. The vertical lines indicate the mean concentrations deduced from the analysis of two filters, the horizontal lines the profile from the real-time data, according to the same graphics scheme as Figure 1.

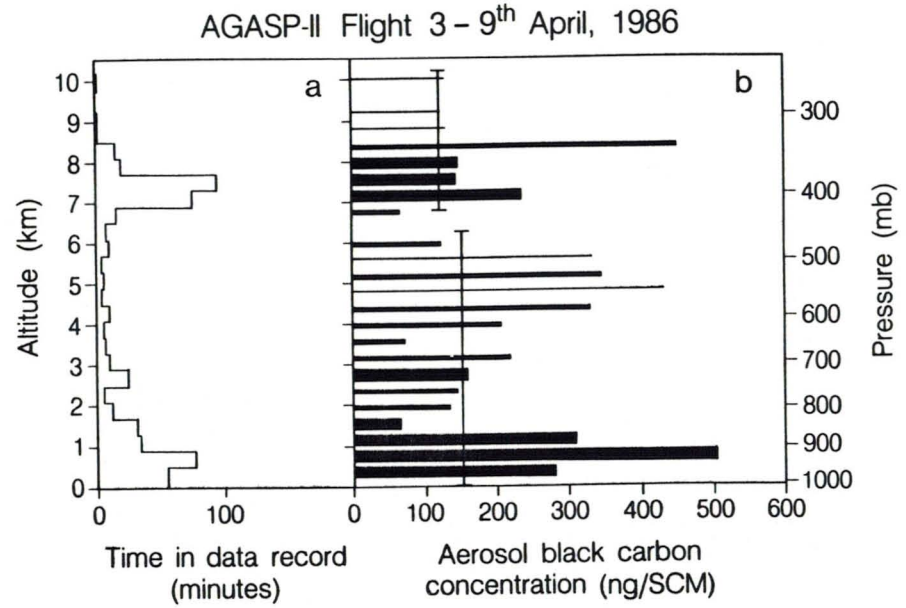


Figure 3. (a) Distribution of data recording time and (b) vertical profile of black carbon concentration averaged for the data set of AGASP-II flight 3. Same graphics scheme as Figure 1

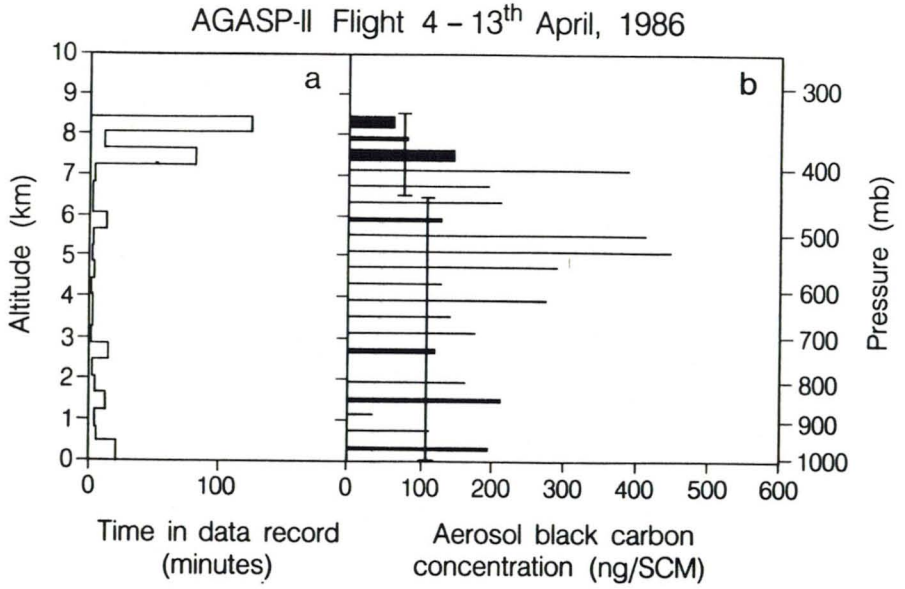


Figure 4. (a) Distribution of data recording time and (b) vertical profile of black carbon concentration averaged for the data set of AGASP-II flight 4. Same graphics scheme as Figure 1.

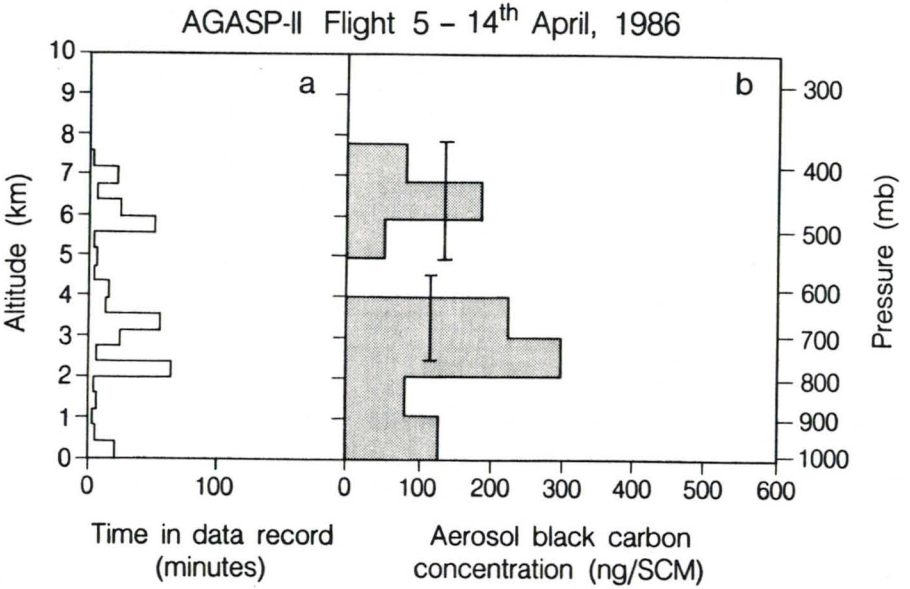


Figure 5. (a) Distribution of data recording time and (b) vertical profile of black carbon concentration averaged for the data set of AGASP-II flight 5. Due to instrumental noise on this flight, the data have been collected into altitude ranges of 1 km extent, and the display format is not weighted by the data collection time.



*Flight 4, April 13, 1986: Alaska-Greenland*

The system recorded data for 321 minutes during this flight, most of which was at high ferry altitudes - two-thirds of the flight at levels above 6.8 km. Only scant coverage of lower altitudes was obtained. The vertical profile of data recording time is shown in Fig. 4(a). The overall mean black carbon concentration was 120 ng $m^{-3}$; however, this value is heavily weighted towards the high-altitude mean of 70 ng $m^{-3}$. Two filters were collected during the flight, one above and one below the 6 km level. The calculated overall vertical profile of black carbon concentration is shown in Fig. 4(b), with the filter results superimposed. Due to the small amount of time spent at lower altitudes, it is not possible to draw any firm conclusions as to the vertical distribution. However, it appears that moderate concentrations were observed at all altitudes.

*Flight 5, April 14, 1986: North Greenland Coast*

During this flight we collected data for 341 minutes. Unfortunately, problems with the aethalometer light source introduced a large amount of noise into the measurements. This noise is noncumulative, and its effects can be reduced by gathering the data into larger time or altitude ranges. We have grouped the data into 1-km altitude range bins. The vertical distribution of data recording time is shown in Fig. 5(a): it indicates reasonably good coverage of the range below 7.5 km with exceptions around 5 km and 1 km altitude. The overall mean black carbon concentration was approximately 200 ng $m^{-3}$. Two filters were collected, covering middle and upper altitude ranges. Their results and a vertical concentration profile of lower resolution are shown in Fig. 5(b). The data generally suggest two layers of aerosol black carbon maxima - one at about 2 km altitude, then a layer of cleaner air at about 4 km, and another polluted layer at about 6 km altitude.

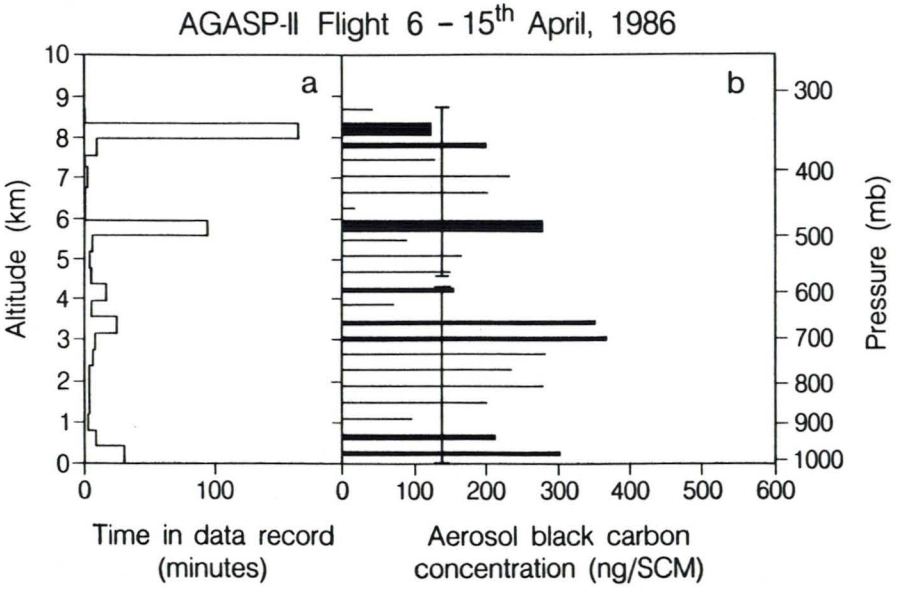


Figure 6. (a) Distribution of data recording time and (b) vertical profile of black carbon concentration averaged for the data set of AGASP-II flight 6. Same graphics scheme as Figure 1.

*Flight 6, April 15, 1986: Greenland-Alaska*

We recorded 411 minutes of data during this flight, much of which was at two ferry altitudes of 5.8 km and 8.3 km. A modest coverage of altitudes below 5.8 km was also obtained. The vertical profile of aethalometer data recording time is shown in Fig. 6(a). We also collected two filters, roughly above and below 4 km altitude. The overall mean black carbon concentration was 205 ng $m^{-3}$; however, this average is heavily biased by the high-altitude data. The overall vertical concentration profile is shown in Fig. 6(b), with the filter results superimposed. Due to the sparse time coverage, it is not possible to determine many details of the profile except for the general appearance of moderate concentrations (200-300 ng $m^{-3}$) below 3.5 km altitude and somewhat reduced concentrations above this altitude, with an indication of a layer of cleaner air in between.

*Composite Profile*

The real-time data from flights 1, 2, 3, 4, and 6 were combined to form a composite profile. The data from flight 5 were excluded due to the instrument noise problem that occurred during that flight. The total amount of data contained in this composite amounts to 2063 measurement minutes distributed over altitude as shown in Fig. 7(a). The coverage of altitude was good except at about 5 km. Substantial amounts of time were spent at lower altitudes (10½ hr below 2 km), and peaks at ferrying altitudes above 7 km are also obvious. The overall composite profile of black carbon is shown in Fig. 7(b). It shows that concentrations aloft are generally higher than at the lowest levels, with a broad maximum of 350 to 500 ng $m^{-3}$ between 0.5 and 2 km altitude. Concentrations at higher altitudes are somewhat lower, averaging around 200 ng $m^{-3}$ from 3 km to 7 km. Even at the highest altitudes, we detected substantial quantities of aerosol black carbon — about 100 ng $m^{-3}$ between 7 km and 8 km, and 25 ng $m^{-3}$ above 9 km altitude.

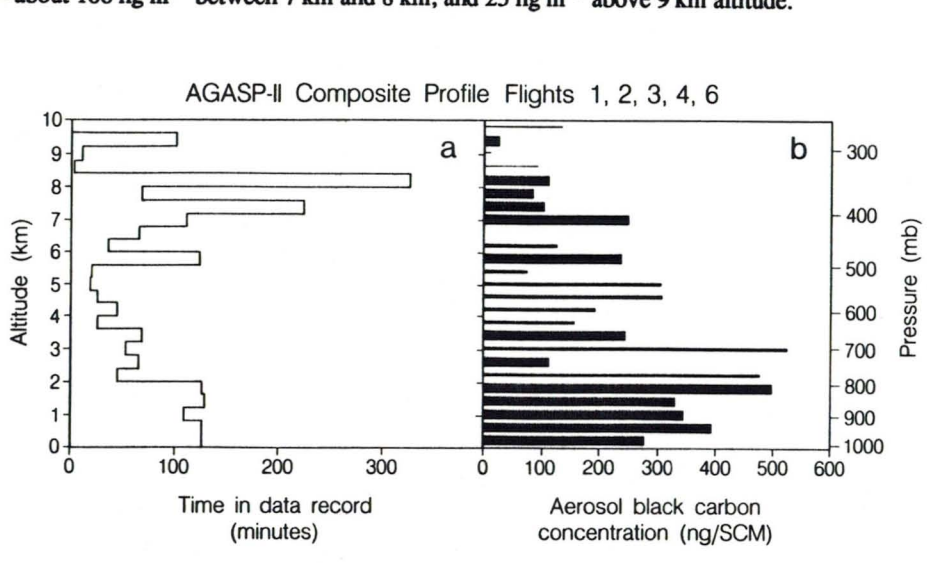


Figure 7. (a) Distribution of data recording time and (b) vertical profile of black carbon concentration for the composite sum of the data sets from AGASP-II flights 1, 2, 3, and 6. The best time coverage was obtained for higher altitudes (6-8 km) and also below 2 km altitude. The profile data are represented by heavy lines for greater than 50 minutes of data recording time per altitude range, and by medium lines for 15 to 50 minutes of data per range.



## 4. Discussion

The highest concentrations of aerosol black carbon were detected at low altitudes during flight 1. A mean value of 585 ng $m^{-3}$ was obtained below 2.5 km altitude. In addition, there was a definite indication of similar concentrations in a layer at about 5 km altitude. There had been some concerns raised as to whether volcanic debris from Mt. St. Augustine was in the sampling zone during this flight. Analysis of the aethalometer filter from flight 1 by temperature-programmed evolved gas analysis for black carbon (Gundel et al., 1984) showed the standard calibration proportionality between the filter analysis and the integral of the real-time output. Thus there was no evidence of substantial interference in the real-time data due to optically absorbing volcanic dust and we conclude that the measured profile indeed represents the vertical distribution of aerosol black carbon. Of course, this carbon may well have been associated with a volcanic plume; further input is necessary from meteorological trajectory analysis and filter sample elemental analysis. We may note, however, that comparable and greater concentrations of aerosol black carbon were detected during AGASP-I in 1983 and that these observations may indicate the long-range transport of combustion emissions of anthropogenic origin. Flight 2, six days later, showed concentrations from 300 ng $m^{-3}$ at low levels to 475 ng $m^{-3}$ at 2.8 km altitude and an indication of a polluted layer between 6 km and 8 km. The following day, flight 3 showed a strong layer near the ground, clean air at about 1.5 km, higher concentrations between 4 km and 5 km, clean air at around 6 km, and then definite concentrations above. Unfortunately there was insufficient sampling time spent in the altitude range 2 km to 6 km to be able to clearly compare the higher altitude data from one day to the next. The low altitude data show a very definite change in the carbon profile from ground level to 3 km altitude over these two days. Flying at 1.5 km altitude (5000 ft) on April 8, the aircraft was in the center of a broad band of aerosol black carbon of concentration approximately 400 ng $m^{-3}$. The following day, April 9, our measurements indicate that this level was a layer of clean air with carbon concentration 70 ng $m^{-3}$. Although these flights were close in time, the profile target zones were separated geographically and the meteorological conditions were quite different. The purpose of this comparison is to show that the haze stratification may be quite variable and that long-range transport does not result in a predictable vertical distribution.

Flights 4 and 6 involved substantial amounts of time spent at high altitudes ferrying between Alaska and Greenland; the profile data received less emphasis but did show definite differences in altitude ranges. Unfortunately we encountered severe instrumental problems on flight 5, limiting the vertical resolution of the recovered data. However, overall profile average concentrations of aerosol black carbon for these flights in the Greenland-Canadian Arctic area were about 200 ng $m^{-3}$, a value close to that determined during flight 3 over the Alaskan North Slope. The high concentrations measured during flight 1 may have been related to the volcanic eruption or may have been due to the passage of a polluted airmass from distant source regions; however, moderate concentrations were detected at all altitudes on all flights in the AGASP-II series.

One possible consequence of the presence of this strongly optically absorbing material suspended in the atmosphere above the highly reflecting polar regions is a perturbation of the solar radiation balance. To estimate the optical effects of the aerosol, it is necessary to measure the vertical profiles of absorption and scattering and to calculate the atmospheric column burdens. Optical scattering is strongly affected by particle size, which in turn may be affected by the interaction of aerosol chemical composition and meteorological history of the polluted airmass - for example the hydration hysteresis of a sulfate-containing submicron particle (Sverdrup and Whitby, 1980; Covert et al., 1980). By contrast, optical absorption by particles in these size ranges is essentially proportional to the volume of absorbing material and is not so greatly affected by changes in par

**Table 1. Mean black carbon concentrations (units nanograms per geometric cubic meter) measured in altitude ranges during AGASP-II.**

| Altitude range | Flight 1 | 2 | 3 | 4 | 5 | 6 | Composite profile |
|---|---|---|---|---|---|---|---|
| 8-10 km | 14 | 80 | 210 | - | - | 120 | 60 |
| 4-8 km | 220 | 120 | 240 | 225 | 110 | 160 | 165 |
| 0-4 km | 435 | 250 | 210 | 160 | 185 | 235 | 335 |

**Table 2. Absorption optical depths (multiplied by 1000) calculated from the concentration data using an absorption cross section of 12 $m^2 g^{-1}$.**

| Altitude range | Flight 1 | 2 | 3 | 4 | 5 | 6 | Composite profile |
|---|---|---|---|---|---|---|---|
| 8-10 km | 0.1 | 0.7 | 1.8 | - | - | 1.0 | 0.5 |
| 4-8 km | 5.3 | 3.0 | 5.9 | 5.5 | 2.7 | 3.9 | 4.0 |
| 0-4 km | 17.5 | 10.0 | 8.5 | 6.4 | 7.4 | 9.4 | 13.3 |
| Column 0-10 km | 23 | 14 | 16 | 12 | 10 | 14 | 18 |

ticle size due to the accretion of transparent species. A representative value for the absorption cross section of a submicron particle containing black carbon mixed with organic and inorganic compounds is 12 $m^2 g^{-1}$ [B.C.] (Faxvog and Roessler, 1978; Yasa et al., 1979). For the six AGASP-II flights and their composite, we can adjust the concentrations to units per geometric cubic meter and calculate the mean concentrations for three altitude ranges as shown in Table 1. Multiplying these concentrations by the column height and this absorption cross section, we can estimate the segment and total column optical depths due to absorption by aerosol black carbon as shown in Table 2.

The total (0-10 km) column absorption optical depths range from 0.01 (flight 5) to 0.023 (flight 1), with a mean value of 0.018. Ambient aerosol single scattering albedos measured in moderately polluted regional source atmospheres range from 0.8 to 0.95 (Japar et al., 1986), implying an accompanying amount of optical scattering from between 4 to 20 times as great as the absorption. Measurements of optical scattering in the Arctic haze have shown values from 5 to $40 \times 10^{-6}$ $m^{-1}$, with calculated single scattering albedos ranging from 0.79 to 0.93 (Clarke et al., 1984) and profiles showing distinct stratification (Wendling et al., 1985). If the carbonaceous aerosols measured during AGASP-II had optical properties falling in the same range, then these measured aerosol black carbon concentrations would contribute values ranging from 0.04 to 0.4 to the total aerosol optical extinction depths. These values are in agreement with total optical depth measurements made at the surface, for example, depths of from 0.2 to 0.5 measured at Barrow, Alaska (Dutton et al., 1984).



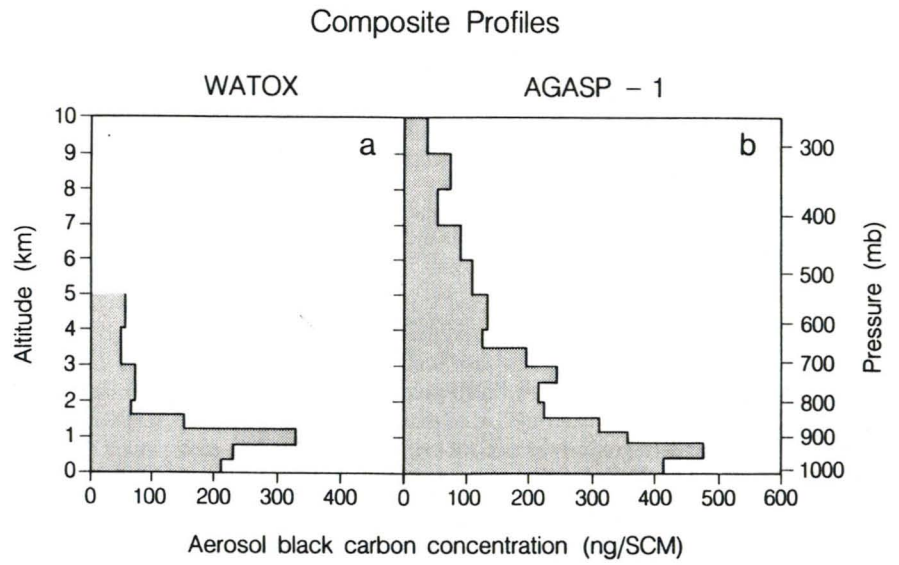


Figure 8. Vertical profiles of aerosol black carbon concentration from other aircraft programs. (a) Composite profile from WATOX-86 (January 1986, western Atlantic Ocean); (b) composite profile from AGASP-I flights 1-7 (March 1983; Alaskan, Canadian, and Greenland Arctic).

The aerosol black carbon concentration measurements may be compared with those from other aircraft missions. Figure 8 shows results from two other programs in which aethalometer measurements were made. Figure 8(a) shows the composite aerosol black carbon profile from the WATOX program flown in January 1986 over the Western Atlantic Ocean (Galloway et al., 1988; Hansen and Novakov, 1988). These flights were generally within 200 km of the east coast of the United States and were timed to follow the eastward passage of a cold front, thus sampling fairly freshly polluted air masses. The highest concentrations of aerosol black carbon were found within the boundary layer; less than 100 ng $m^{-3}$ were measured above the boundary up to the operational ceiling of 5 km altitude. In all measurements, these concentrations were less than those measured at similar altitudes in the Arctic haze. Figure 8(b) shows the composite aerosol black carbon profile for flights 1-7 of the 1983 AGASP-I program, namely flights in the same general areas of Alaska, Canada, and Greenland as covered by AGASP-II. Although peak carbon concentrations measured within polluted strata during AGASP-I exceeded those measured during AGASP-II, comparison of Fig. 8(b) with Fig. 7(b) shows that the mean concentrations were in general higher during AGASP-II than AGASP-I. This observation is supported by comparison of the total tropospheric column absorption optical depths calculated from the carbon measurements for these two Arctic programs. During AGASP-I, column (0-10 km) absorption optical depths ranged from 10 to $17 \times 10^{-3}$, with the composite profile (Fig. 8(b)) yielding a value of $13 \times 10^{-3}$. These figures may be compared with those in the lowest row of Table 2. From this we may conclude that the carbon concentrations measured aloft during the two AGASP missions were not due to unusual or isolated circumstances, but that substantial concentrations of this combustion-derived pollutant species may exist throughout the Arctic troposphere during the winter and spring Arctic haze.

The equivalent mean tropospheric column burden of aerosol black carbon deduced from the AGASP measurements is on the order of 1-1.5 mg $m^{-2}$. The deposition velocity to the surface for this aerosol species in the stratified winter Arctic atmosphere has not yet been determined. However, we may estimate the total deposition of aerosol black carbon to the surface during the winter and spring seasons as lying in the range from 1 to 10 mg $m^{-2}$. Multiplying this amount by the absorption cross section of the aerosol (approximately 12 $m^2 gram^{-1}$) shows that the effect of this deposit in changing the albedo of a snow or ice surface may be considerable. Direct measurements of the elemental carbon content of Arctic snowpacks and calculations of the radiative-transfer effects have been discussed by, for example, Clarke and Noone (1985) and Warren (1984).

A typical urban mix of combustion sources in the United States has an overall emission factor in the range of 1 to $5 \times 10^{-4}$ for the fraction of carbon atoms in the fuel that are emitted in the particulate phase as aerosol black carbon. For less well-regulated combustion, this fraction may increase by a factor of 10. A combustion effluent plume therefore contains three to four orders of magnitude more carbon emitted as carbon dioxide than as particulate material, and we could anticipate that the measured concentrations of aerosol black carbon may be accompanied by increases in carbon dioxide concentration of a few parts per million. Observations of increased carbon dioxide have been reported for season-long ground-based measurements (Halter et al., 1985), airborne measurements (Conway et al., 1985; Conway et al., this issue), and correlations of $CO_2$ with aerosol black carbon have been measured in real-time plume impact measurements at ground level (Hansen et al., this issue). In conjunction with meteorological analysis, these results suggest that the observed aerosol black carbon concentrations in the Arctic haze are due to combustion emissions from large-scale distant source regions.

Finally, we note that the aerosol black carbon concentrations measured at the highest operational altitudes were definite and measurable, in the range of 25-50 ng $m^{-3}$. Ozone measurements at these altitudes started to rise at times, indicating that the flight level was sampling at least partially mixed stratospheric air. This mixing might result in the injection of some aerosol black carbon from the upper troposphere to the lower stratosphere, (Chuan and Woods, 1984) with possible physical and chemical consequences. Recent laboratory studies of the reactions between the carbon surface and ozone (Sergides et al., 1987; Stephens et al., 1986) have shown both catalytic ozone-destroying and carbon-consuming characteristics. Even if carbon is used only as a tracer for combustion emissions, its presence in an airmass is an indicator of the impact of emissions on that airmass with the probable accompaniment of other pollutant species.

## 5. Conclusions

During the second Arctic Gas and Aerosol Sampling Program, we performed airborne measurement of aerosol black carbon from ground level to 10 km altitude. Typical concentrations at low altitudes were 300-500 nanograms per standard cubic meter, decreasing gradually to about 100 ng $m^{-3}$ at 7-8 km altitude. Stratification of the polluted airmasses was observed on some flights, and large variations in the vertical profile were measured. Mean concentrations were generally slightly higher than those measured during AGASP-I. These concentrations could lead to perturbations of the solar radiation balance, either due to an increased optical depth in the atmospheric column or due to deposition on the surface. The candidate sources are probably distant regions of considerable fuel consumption. The detection of particulate carbon in the upper troposphere may imply that it is mixing into the stratosphere.



## 6. Acknowledgments

The authors wish to acknowledge the technical expertise of Mr. R.C. Schmidt in constructing essential elements of the equipment. We thank Dr. R.C. Schnell (CIRES, Boulder, Colorado) for coordinating the aircraft program, and the NOAA Office of Aircraft Operations, their pilots, engineers, and support staff for their excellent field operation. Funding for this work was provided by the U.S. Department of Energy under contract DE-AC03-76SF00098, by the National Oceanic and Atmospheric Administration under contract 40RAN520248, and by the Defense Nuclear Agency under contract BR 0330 1000.

## 7. References

Andreae, M.O. (1983) "Soot carbon and excess fine potassium: Long-range transport of combustion-derived aerosols," Science **220**, 1148-1151.

Barrie, L.A. (1986) "Arctic air pollution: An overview of current knowledge" Atmos. Environ. **20**, 643-663.

Bizjak, M., V. Hudnik, A.D.A. Hansen, and T. Novakov (1986) "Evidence for heterogeneous $SO_2$ oxidation in Ljubljana, Yugoslavia," Atmos. Environ. **20**, 2199-2204.

Brodzinsky, R., S.-G. Chang, S.S. Markowitz, and T. Novakov (1980) "Kinetics and mechanism for the catalytic oxidation of sulfur dioxide on carbon in aqueous suspensions," J. Phys. Chem. **84**, 3354-3359.

Cess, R.D. (1983) "Arctic aerosols: Model estimates of interactive influences upon the surface-troposphere radiation budget," Atmos. Environ. **17**, 2555-2564.

Chuan, R.L., and D.C. Woods (1984) "The appearance of carbon aerosol particles in the lower stratosphere," Geophys. Res. Lett. **11**, 553-556.

Clarke, A.D., and K.J. Noone (1985) "Soot in the Arctic snowpack: A cause for perturbations in radiative transfer," Atmos. Environ. **19**, 2045-2053.

Clarke, A.D., R.J. Charlson, and L.F. Radke (1984) "Airborne observations of Arctic aerosol: Optical properties of Arctic haze," Geophys. Res. Lett. **11**, 405-408.

Coakley, J.A., R.D. Cess, and F.B. Yurevich (1983) "The effect of tropospheric aerosols on the earth's radiation budget: A parameterization of climate models," J. Atmos. Sci. **116**, 137-144.

Conway, T.J., and L.P. Steele (1989) "Carbon dioxide and methane in the Arctic atmosphere," J. Atmos. Chem., this issue.

Conway, T.J., W.E. Raatz, and R.H. Gammon (1985) "Airborne $CO_2$ measurements in the Arctic during spring 1983," Atmos. Environ. **19**, 2195-2202.

Covert, D.S., A.P. Waggoner, R.E. Weiss, N.C. Ahlquist, and R.J. Charlson (1980) "Atmospheric aerosols, humidity and visibility," in *The Character and Origins of Smog Aerosols*, G.M. Hidy et al., eds. (New York, Wiley), pp. 559-581.

Dutton, E.G., J.J. DeLuisi, and B.A. Bodhaine (1984) "Features of aerosol optical depth observed at Barrow, March 10-20, 1983," Geophys. Res. Lett. **11**, 385-388.

Faxvog, F.R., and D.M. Roessler (1978) "Carbon aerosol visibility vs. particle size distribution," Appl. Opt. **17**, 2612-2616.

Galloway, J.N., ed. (1988) *WATOX-86 Special Issues*, Global Biogeochem. Cycles **1**, 261-380; **2**, 1-71.

Groblicki, P.J., S.H. Cadle, C.C. Ang, and P.A. Mulawa (1983) *Interlaboratory comparison of methods for the analysis of organic and elemental carbon in atmospheric particulate matter*, Research Publication GMR-4054-ENV-152, General Motors Research Laboratories, Warren, Michigan.

Gundel, L.A., R.L. Dod, H. Rosen, and T. Novakov (1984) "The relationship between optical attenuation and black carbon concentration for ambient and source particles," Sci. Total Environ. **36**, 197-202.

Halter, B.C., J.M. Harris, and K.A. Rahn (1985) "A study of winter variability in carbon dioxide and Arctic haze aerosols at Barrow, Alaska," Atmos. Environ. **19**, 2033-2037.

Hansen, A.D.A., T.J. Conway, L.P. Steele, B.A. Bodhaine, K.W. Thoning, P. Tans, and T. Novakov (1989) "Correlations among combustion effluent species at Barrow, Alaska: Aerosol black carbon, carbon dioxide, and methane," J. Atmos. Chem., this issue.

Hansen, A.D.A., and T. Novakov (1988) "Aerosol black carbon measurements over the western Atlantic Ocean," Global Biogeochem. Cycles **2**, 41-45.

Hansen, A.D.A., B.A. Bodhaine, E.G. Dutton, and R.C. Schnell (1988) "Aerosol black carbon measurements at the South Pole: Initial results, 1986-1987," submitted to Geophys. Res. Lett.

Hansen, A.D.A., H. Rosen, and T. Novakov (1982) "Real-time measurement of the absorption coefficient of aerosol particles," Appl. Opt. **21**, 3060-3062.

Japar, S.M., W.W. Brachaczek, R.A. Gorse, Jr., J.M. Norbeck, and W.R. Pierson (1986) "The contribution of elemental carbon to the optical properties of rural atmospheric aerosols", Atmos. Environ. **20**, 1281-1289.

Marland, G., R.M. Rotty, and N.L. Treat (1985) "$CO_2$ from fossil fuel burning: Global distribution of emissions," Tellus **37B**, 243-258.

Penner, J.E., and L.L. Edwards (1986) "Nucleation scavenging of smoke and aerosol particles in convective updrafts," in *Proceedings, 23rd Conference on Radar Meteorology and Conference on Cloud Physics, Snowmass, Colorado, September 22-26, 1986*, v. 2 (American Meteorological Society), pp. 83-86.

Pueschel, R.F., E.W. Barrett, D.L. Wellman, and J.A. McGuire (1981) "Cloud modification by man-made pollutants: Effects of a coal-fired power plant on cloud drop spectra," Geophys. Res. Lett. **8**, 221-224.

Rahn, K.A., ed. (1981) *Arctic Air Chemistry Special Issue*, Atmos. Environ. **15**, 1345-1516.

Rahn, K.A., ed. (1985) *Arctic Air Chemistry Special Issue*, Atmos. Environ., **19**, 1987-2207.

Rosen, H., and A.D.A. Hansen (1985) "Estimates of springtime soot and sulfur fluxes entering the Arctic troposphere: Implications to source regions," Atmos. Environ. **19**, 2203-2207.

Rosen, H., A.D.A. Hansen, L.A. Gundel, and T. Novakov (1978) "Identification of the optically absorbing component of urban aerosol particles," Appl. Opt. **17**, 3859-3861.

Schnell, R.C., ed. (1984) *Arctic Haze Special Issue*, Geophys. Res. Lett. **11**, 359-472.

Sergides, C.A., J.A. Jassim, A.R. Chughtai, and D.M. Smith (1987) "The structure of hexane soot. 3 - Ozonation studies," Appl. Spectrosc. **41**, 482-492.

Shaw, G.E., and K. Stamnes (1980) "Arctic haze: Perturbations of the polar radiation budget," Ann. N.Y. Acad. Sci. **338**, 533-539.



Stephens, S., M.J. Rossi, and D.M. Golden (1986) "The heterogenesis reaction of ozone on car bonaceous surfaces," Int. J. Chem. Kinetics **18**, 1133-1149.

Sverdrup, G.M., and K.T. Whitby (1980) "The effect of changing relative humidity on aeroso size distribution measurements," in *The Character and Origins of Smog Aerosols*, G.M Hidy et al., eds. (New York, Wiley), pp. 527-558.

Twomey, S. (1977) "The influence of pollution on the shortwave albedo of clouds," J. Atmos Sci. **34**, 1149-1152.

Valero, F.P.J., T.P. Ackerman, and W.J.Y. Gore (1984) "The absorption of solar radiation by th Arctic atmosphere during the haze season and its effects on the radiation balance," Geo phys. Res. Lett. **11**, 465-468.

Warren, S.G. (1984) "Impurities in snow: Effects on albedo and snowmelt (Review)," Annal Glaciol. **5**, 177-179.

Wendling, P., R. Wendling, W. Renger, D.S. Covert, J. Heintzenberg, and P. Moerl (1985) "Cal cualted radiative effects of Arctic haze during a pollution episode in spring 1983 based o ground-based and airborne measurements," Atmos. Environ. **19**, 2181-2193.

Wolff, G.T. (1984) "Particulate elemental carbon in the atmosphere," JAPCA **31**, 935-938, an references therein.

Yasa, Z., N.M. Amer, H. Rosen, A.D.A. Hansen, and T. Novakov (1979) "Photoacoustic investi gations of urban aerosol particles," Appl Opt. **18**, 2528-2530.

(Received 7 September 1987)