

HORIZONTAL INHOMOGENEITIES IN THE PARTICULATE CARBON COMPONENT OF THE ARCTIC HAZE

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Abstract—We present results obtained during the 'AGASP 1983' Arctic haze aircraft sampling experiment. During this program we operated the aethalometer, an instrument that responds to the aerosol graphitic ('black') carbon concentration in real time. Previous results showed strong vertical layering of this component of the Arctic haze. In this paper we present for the first time evidence of horizontal variations of aerosol black carbon concentration. Some of these variations correlate with meteorological parameters, but we also observed horizontal inhomogeneities with a characteristic scale of 50–100 km occurring in the absence of meteorological activity.

Key word Index: Arctic haze, aerosols—optical absorption by, AGASP 1983 experiment, airborne measurements, elemental particulate carbon, distribution of aerosol—inhomogeneous, vertical, horizontal, long-range transport, soot—use as air mass tracer.

INTRODUCTION

The Arctic haze has been found to have a large carbonaceous component derived from combustion emissions, with submicrometer particles containing both organic compounds and microcrystalline graphitic forms (Rosen and Novakov, 1983). This latter species is often termed 'black carbon' due to its large optical absorption cross-section. It can only be produced during incomplete combustion and is inert to transformation in the atmosphere. There are therefore no sources of black carbon within the atmosphere, making this material a good tracer for the evolution of a polluted air mass. Particulate black carbon was found throughout the Arctic troposphere during the AGASP 1983 program in sufficient concentrations that its optical absorption may lead to a significant perturbation of the solar radiation balance (Shaw and Stamnes, 1980; Pörr and MacCracken, 1982; Cess, 1983; Rosen and Hansen, 1984; Valero *et al.*, 1984). Knowledge of the distribution of this material is necessary for these calculations, and the use of black carbon as an air mass tracer may improve our understanding of transport processes. We have previously reported results on the vertical distributions of aerosol black carbon in the Arctic haze, showing strong layering structure as well as significant concentrations at all altitudes in the troposphere (Hansen and Rosen, 1984; Rosen and Hansen, 1984). In this paper we present results from detailed analyses of portions of the flight tracks showing substantial horizontal variations of black carbon concentration, often (but not always) in conjunction with meteorological features.

EXPERIMENTAL DETAILS

Our installation aboard the AGASP WP-3D aircraft included the aethalometer, an instrument developed at Lawrence Berkeley Laboratory that collects the aerosol on a filter and uses a continuous optical technique to measure the concentration of aerosol black carbon in real time (Hansen *et al.*, 1982). The filter is changed periodically to yield an integrated sample available for chemical analysis. Methods for the optical and chemical determination of microgram quantities of black carbon are well developed (Gundel *et al.*, 1984), and their results when applied to these filters are used to calibrate the aethalometer. From the data reduction routines, we obtained a set of black carbon concentrations, averaged over 1-min intervals and expressed in units of nanograms of aerosol black carbon per geometric cubic meter. Note that these units, necessary for the calculation of aerosol optical properties, are not altitude independent as is a mixing ratio. The estimated error on the measurements presented here is $\pm 0.09 \mu\text{g m}^{-3}$. We then meshed these aethalometer data with the aircraft systems tape to give a data base containing average readings every minute for black carbon concentration, position and altitude, wind speed and direction, air temperature and relative humidity, etc. (Schnell, 1984). The periods of time during which all the above measurements were valid add to a total of approximately 70 h of flying time north of 70° latitude.

The flight patterns employed during the AGASP 1983 program included both rapid and gradual descents and ascents, as well as portions of level flight paths at various altitudes down to 30 m ASL. We scanned the data base to search for flight track portions that indicated a horizontal pass through a varying black carbon concentration. Although the AGASP flight patterns were not designed to study horizontal inhomogeneities, we found several suitable events in this scan. Details from three of these are presented below.

RESULTS

Figure 1 shows data obtained during AGASP flight 9 over a period of approximately 35 min during which the aircraft flew in a constant northerly track at altitudes between 83 m and 91 m ASL. The track covered a distance of approximately 250 km from 75.55° N, 26.2° E to 78.22° N, 27.2° E, i.e. somewhat to the east of Spitzbergen. From zero to 50 km on the track scale, the aircraft was over ocean with broken ice; from 50 km onwards, the surface was more or less continuous ice with leads. The air temperature had been steady at around 270 K before the ice was reached; it then gradually decreased to 265 K at the 200-km mark. During this time, the wind direction was steady at 200–210°, and the dew point was approximately constant at 261 K; i.e. the relative humidity increased from 60% to 80%. These data suggest that the aircraft was flying in a single moist air mass whose temperature was dropping. Conditions were noted as being overcast and visibly hazy. The black carbon concentration increased from about $0.25 \mu\text{g m}^{-3}$ to a peak of $1.6 \mu\text{g m}^{-3}$ at the 200-km mark. At this point, the observer's record states that snow flurries had started and continued for a few minutes until by the 250-km mark the aircraft had flown through a front into clear skies. The wind direction shifted by 55°, the air temperature fell by 5 K, and the dew point fell by 10°. The new air mass was colder and drier, and its black carbon concentration was down to about $0.25 \mu\text{g m}^{-3}$.

Figure 2 shows 47 min of data from AGASP flight 3,

encompassing almost a complete loop of length 130 km and height from 30 m to 1800 m altitude. The descent spiral was at 72.7° N, 156.8° W and the ascent was a climb at 71.5° N, 157.4° W. The low-level portion was a southerly track from about 200 km to 50 km north of Barrow. During this portion, the wind speed and direction were steady at $4.3 \pm 0.4 \text{ m s}^{-1}$ and $51 \pm 8^\circ$, respectively, and the relative humidity was $75 \pm 5\%$. There was no clear evidence of a changing air mass. However, Fig. 2a shows that the average black carbon concentration increased steadily from about $0.25 \mu\text{g m}^{-3}$ to almost $1 \mu\text{g m}^{-3}$ along the low-level track portion; and Fig. 2b shows that the air temperature increased by 4 K in the same period. At the end of this track portion, the aircraft climbed and essentially retraced its track at 1850 m altitude for about 100 km distance. We have constructed contours of aerosol black carbon concentration for the purposes of visualization, based on interpolation of the data points around the track perimeter. We emphasize that these are not measured contours; however, because there were neither sources nor sinks of aerosol black carbon within this perimeter, the implied continuity is reasonable. The data therefore suggest a concentration of black carbon at low altitudes, with a maximum at some point beyond the 150-km track point. However, there was no evidence of precipitation nor crossing of an air mass boundary during the flight loop.

Figure 3 shows 51 min of data taken during AGASP flight 5. This track portion was a straight path from 71.8° N, 141.4° W to 74.0° N, 137.3° W over the Beaufort Sea during the flight from Anchorage,

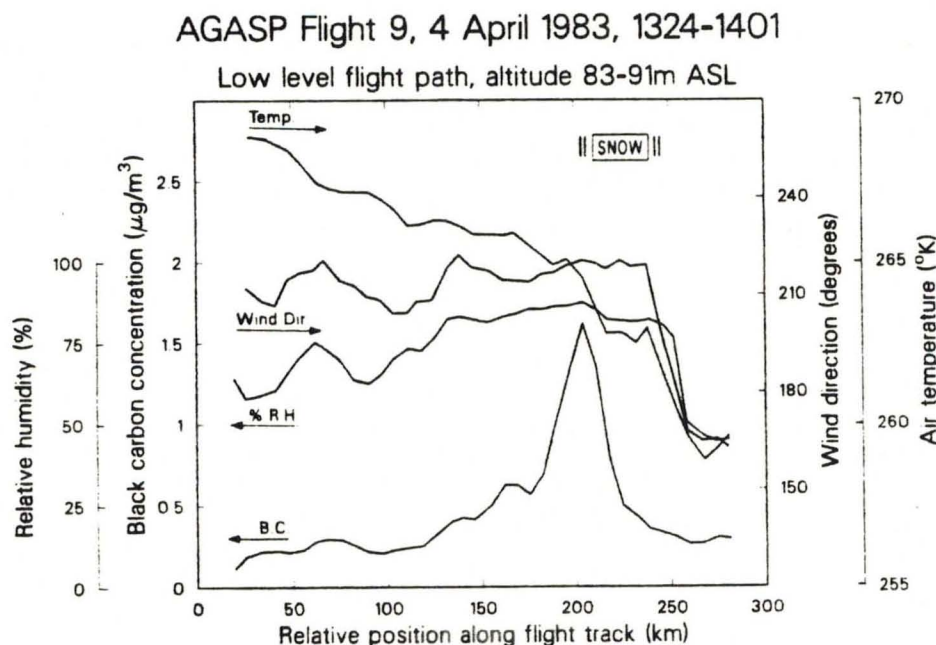


Fig. 1. Data taken during low-level flight from 1324 to 1401 UT, 4 April 1983. Air temperature and wind direction scales are on the right-hand axis; relative humidity and aerosol black carbon concentration scales are on the left. The track was straight and northerly with the horizontal origin at 75.5° N, 26.2° E. The bar at upper right indicates the observation of snow flurries.

AGASP Flight 3, 15 March 1983, 2304-2351

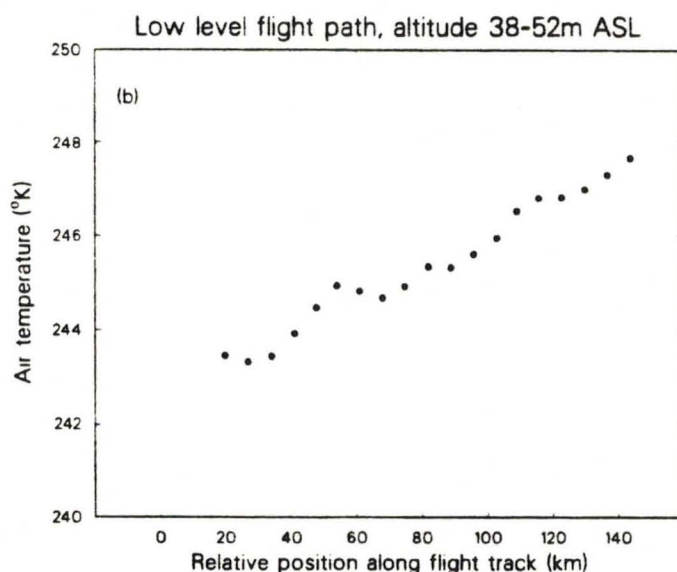
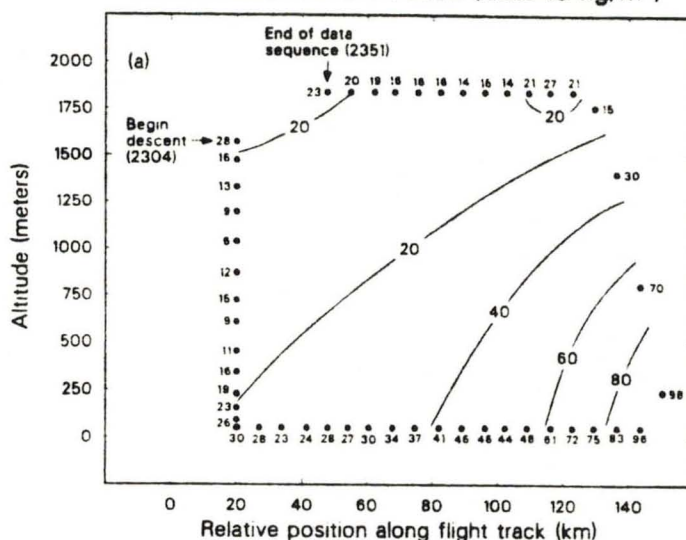
Black carbon concentration (units 10 ng/m^3)

Fig. 2. Data taken during loop flight pattern from 2304 to 2351 UT, 15 March 1983. The descent was a spiral at 72.7°N , 156.8°W ; the level flight track was southerly. (a) Aerosol black carbon concentration vs altitude and relative horizontal position. The contours are hypothetical, calculated by interpolation of the perimeter measurements. (b) Air temperature for the low-altitude flight track portion from 22317 to 2335 UT.

Alaska, to Thule Air Base, Greenland. The Canadian coast was from about 100 to 400 km distant during this time. The data represent a descent to 30 m altitude followed by about 17 min of flying at low altitudes, after which the aircraft climbed to 1550 m ASL altitude. Figure 3b shows the air temperature and wind direction. A strong temperature inversion is seen, with a 10 K fall between 700 m altitude and sea level. The wind direction shifted very little along the first 250 km

of the track but then rapidly turned by 50° soon after the climb-out. At this time, although the air temperature showed little change, the dew point rose from 237 K to 252 K, indicating that the aircraft had entered a different air mass. The black carbon data presented in Fig. 3a show a concentration of the order of $0.2 \mu\text{g m}^{-3}$ at high altitudes at the start of the sequence. This increased to $0.73 \mu\text{g m}^{-3}$ at about 800 m altitude on the descent portion and then fell off rapidly

AGASP Flight 5, 21 March 1983, 1407-1458

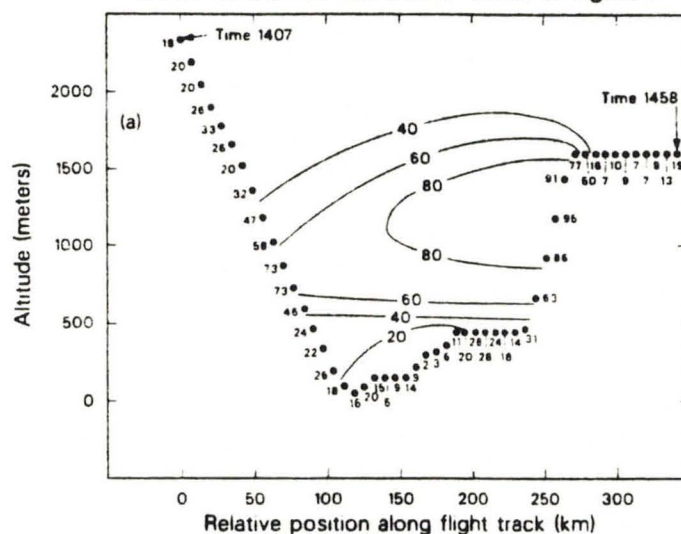
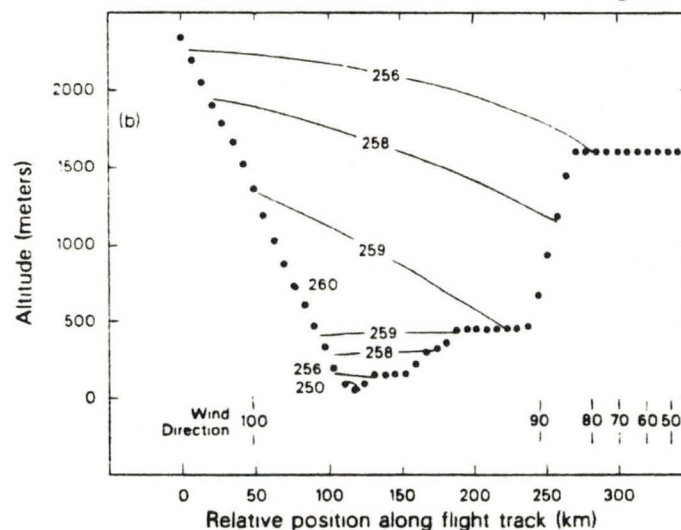
Black carbon concentration (units 10 ng/m^3)Air temperature ($^{\circ}\text{K}$) and wind direction (degrees)

Fig. 3. Data taken during descent/ascent flight pattern from 1407 to 1458 UT, 21 March 1983. The track was straight and northeasterly with the horizontal origin at 71.8°N , 141.4°W . (a) Aerosol black carbon concentration vs altitude and horizontal position. (b) Air temperature and wind direction vs altitude and position. In both figures the contours are estimated from the perimeter measurements and drawn for visualization purposes only.

to low values at low altitudes when the aircraft was within the surface temperature inversion air layer. Upon climbing, the black carbon concentration increased again to almost $1 \mu\text{g m}^{-3}$ but then rapidly disappeared at the start of the level flight portion at the same time as the changing meteorological measurements indicated an air mass boundary. The constructed contours shown in the figure suggest a concentration maximum close to this point.

DISCUSSION

The data presented in Fig. 1 suggest the following situation, similar to one described by Raatz and Schnell (1984) for another AGASP flight in the Norwegian Arctic. The aircraft was flying on a northerly track at a constant low altitude in a moist, polluted air mass. The black carbon concentration started increasing substantially at around the 150-km

track mark but fell rapidly as soon as precipitation started. Shortly thereafter, the aircraft passed through a front into an air mass whose aerosol black carbon concentration was low. The removal of aerosols by precipitation is expected; however, there was no ongoing meteorological activity to explain the five-fold change in black carbon concentration between the start of the track and the peak at the 200-km mark. Figure 2a shows a similar situation, in which the aerosol black carbon concentration increased four-fold within the same air mass. Here, the air temperature gradually rose (see Fig. 2b), but there was no clear evidence of recent meteorological activity or the approach of a front.

There may be several possible explanations for these observations. One is that the air masses were at one time uniformly polluted but that precipitation or other scavenging had occurred at some time in the past over only part of the air mass, removing aerosols from a portion of the flight track. Another is that the injection of carbonaceous aerosol into the air mass had been confined in an area at the source region and that these are plume edge profiles. However, this would require only a small amount of horizontal dispersion over the long transport paths (see Harris, 1984) and imply a diffusion coefficient of the order of $10^9 \text{ cm}^2 \text{ s}^{-1}$. At present, we cannot unambiguously identify either these or other possible mechanisms as explanation for the data.

The maximum instantaneous concentration of black carbon observed during the entire AGASP program was approximately $1.5 \mu\text{g m}^{-3}$, which we shall take as a reasonable value for the maximum concentration likely to be found at the center of an inhomogeneity. If we assume that the flight track of Fig. 2a penetrated the edge of such a patch, we can estimate the size scale of the patch from the spacing of the contours. The result of applying these projections suggests a horizontal scale of 50–100 km as the characteristic length over which the aerosol black carbon concentration changes by a factor of two. We must strongly emphasize that this distance represents a characteristic length on the basis of partial perimeter measurements, rather than an actual complete mapping. The data shown in Fig. 1 from the start of the track to the onset of precipitation also fit into this scale.

The data shown in Fig. 3 present a more complex situation. Figure 3b indicates a surface temperature inversion layer extending to about 400 m altitude and an air mass boundary at about 280 km on the track. The constructed contours in Fig. 3a show strong aerosol black carbon concentrations in the air above the surface layer but very low levels below the inversion, an observation in agreement with the expected weak coupling of these layers. The effect of the front at 280–300 km track is similar to that shown in Fig. 1: in the new air mass, the black carbon concentrations are low. The constructed contours indicate a very rapid change of aerosol black carbon concentration at the front and also show that the polluted air mass is resting

on top of a clean surface temperature inversion layer. This contrasts with the situation often found in urban atmospheres, in which the emissions are trapped in the surface layer beneath the inversion. This observation in the Arctic may imply long-range transport of combustion emissions (Rahn and McCaffrey, 1980; Iversen, 1984).

CONCLUSION

During the AGASP 1983 flight program, we obtained real-time measurements of the black carbon component of the Arctic haze. We examined this data base and found a few examples of horizontal inhomogeneities in the concentration of this material. Some of these effects were due to precipitation or the passing of fronts; however, we also observed changes of concentration at constant altitude in the absence of ongoing meteorological activity. In these instances, the aerosol black carbon appeared to be found in patches with a characteristic horizontal scale of the order of 50–100 km. At present, we cannot determine the mechanisms that lead to this structure. Candidates include inhomogeneous injection and transport, and inhomogeneous removal from a larger, uniformly polluted air mass. These results are based on events fortuitously included in the data base of the multipurpose AGASP 1983 experiment, and indicate the need for further measurements. The ability to measure aerosol black carbon quantitatively in real time from an aircraft will assist studies of its origins, transport, effects and dispersal.

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