

Soot in the Atmosphere and Snow Surface of Antarctica

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Samples of snow collected near the south pole during January and February 1986 were analyzed for the presence of light-absorbing particles by passing the melted snow through a nuclepore filter. Transmission of light through the filter showed that snow far from the station contains the equivalent of 0.1–0.3 ng of carbon per gram of snow (ng/g). Samples of ambient air were filtered and found to contain about 1–2 ng of carbon per kilogram of air, giving a scavenging ratio of about 150. The snow downwind of the station exhibited a well-defined plume of soot due to the burning of diesel fuel, but even in the center of the plume 1 km downwind, the soot concentration was only 3 ng/g, too small to affect snow albedo significantly. Measurements of snow albedo near large inland stations are therefore probably representative of their surrounding regions.

INTRODUCTION

A model of radiative transfer in snow was used by *Wiscombe and Warren* [1980] to explain the reasons for the observed variability of snow albedo. Snow grain size was found to be the most important variable controlling the albedo, and the normal growth of snow grains by metamorphism was sufficient to explain observed variations of spectral albedo in the near-infrared, where the albedo is low. At visible wavelengths, however, where the albedo is high, the measured albedo was not as high as the model predicted, and could not be explained by the model for any realistic grain sizes, so *Warren and Wiscombe* [1980] concluded that the snow in these (northern-hemisphere) experiments was not pure. The variation of absorption with wavelength indicated that the most likely absorptive impurity was graphitic carbon. This idea was confirmed by *Grenfell et al.* [1981], who measured snow albedo in the Cascade Mountains and then analyzed the snow for its soot content, essentially validating the model's prediction for a mixture of soot in snow [*Warren*, 1982].

Antarctic snow should be much cleaner than northern-hemisphere snow, so it was puzzling that the only measurement of Antarctic snow albedo with good spectral resolution [*Kuhn and Siogas*, 1978] (reproduced as Figure 2.2 of *Schwerdtfeger* [1984]) also showed rather low values in the visible. These were likely due to experimental errors (M. Kuhn, personal communication, 1984), but another possible explanation, favored by *Warren and Wiscombe* [1980] was that they were made too close to the station and therefore subject to local pollution. This raised the possibility that radiation measurements made near large inland antarctic stations might not be representative of the surrounding plateau. To resolve this question a field experiment was carried out at the south pole December 1985 to February 1986 by T. C. Grenfell, S. G. Warren, and P. C. Mullen [*Warren et al.*, 1986] to measure optical properties of snow as well as the absorptive impurities in the snow.

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The wind at the south pole almost always comes from the sector 40°W to 100°E [*Robinson et al.*, 1988, Figure 2], i.e., on average somewhat to the left of the topographic gradient. When the wind does blow from other directions it is weak. The airport runway (Figure 1) is oriented close to the prevailing wind direction (020°). (All directions given in this paper are on the local Antarctic grid, where "north" (000°) is parallel to the Greenwich meridian in the direction of Greenwich.)

EXPERIMENT

Snow samples were collected along lines extending radially from the station in the directions 020°, 110° and 290°. Beginning at the limits of the main station activities, samples were collected at distances 0.1, 0.2, 0.5, 1.0, and 2.0 km along these lines (Figure 1), as determined by a hand-pushed bicycle-wheel odometer. Downwind of the station, samples were collected along three lines running in the 110°–290° direction, so as to obtain transects of the expected plume of pollution. These lines were at distances 0.4, 1.2, and 2.4 km downwind from the station, the farthest line located just downwind of the start of the airport runway (Figure 1).

The design of the sampling strategy was facilitated by our being able to carry out preliminary soot analyses at the station within a day or so after collecting the snow, so that subsequent sampling locations could be chosen on the basis of these analyses. Following the method developed by *Clarke and Noone* [1985], snow samples were collected in glass jars, melted in a microwave oven and quickly passed through a 0.4- μm nuclepore filter into an Erlenmeyer flask with partial vacuum provided by a hand pump. The filters were compared visually to four standard filters which contained known amounts of soot. The filters were later analyzed accurately with a 4-wavelength photometer at the University of Hawaii, but the preliminary estimate by eye was sufficient to choose the locations of further samples to be collected. Over 100 samples were extracted and analyzed at the station. Duplicates of a few samples were kept frozen for subsequent analysis at Seattle and Hawaii.

The soot content of snow near the station varies greatly on

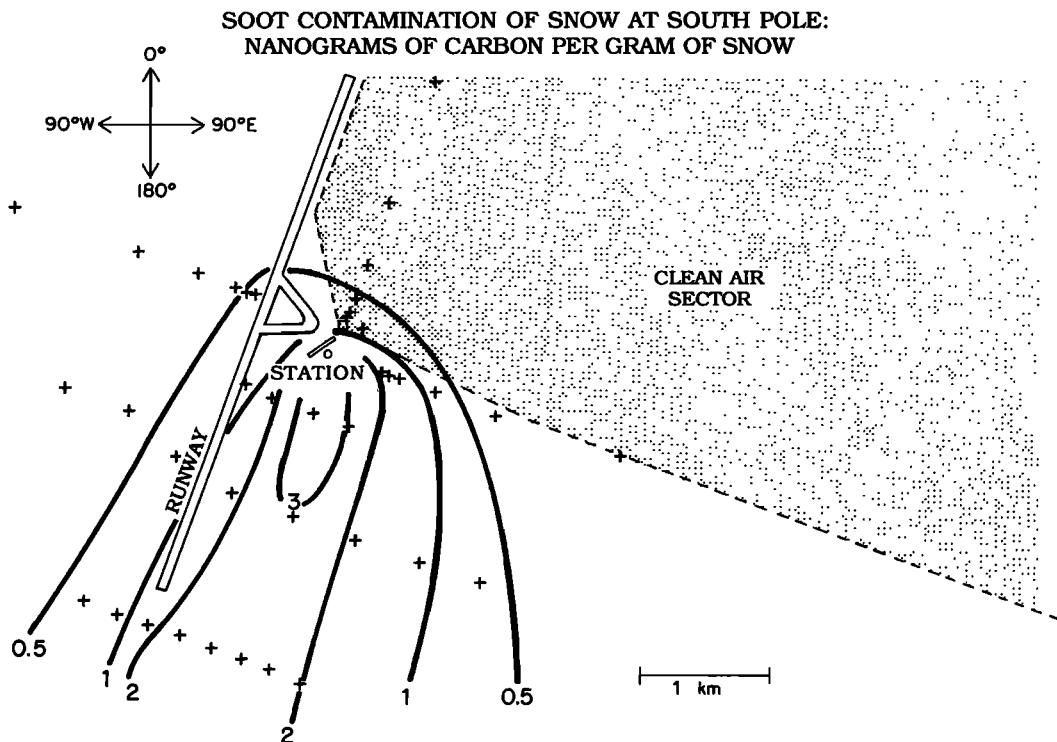


Fig. 1. Contamination of snow by soot at Amundsen-Scott South Pole Station. Values are given in nanograms of carbon per gram of snow. Sampling sites are indicated by plus signs. All samples used to draw the contour lines were taken from sastrugi in order to represent approximate upper limits to the EC content. The contour lines of equal EC content are only approximate because of scatter in the data values.

horizontal scales of less than 1 meter, because of surface structures in the snow. The snow surface at the end of winter consists of hard snow ridges ("sastrugi") formed by wind erosion, about 0.5 m high and oriented with the strong winds. The sastrugi decay somewhat by sublimation during the summer, as explained by Gow [1965], but remnants of them remain, which would retain the soot left behind after sublimation. The snow in sastrugi downwind of the station has potentially been exposed to pollution of many months, and is normally more contaminated (by as much as a factor of 10) than the loose snow between the sastrugi which may have been carried into the region of the station by a blizzard just 1 day before we collected the samples.

All the snow samples used to draw the contour lines in Figure 1 were taken from old hard sastrugi, so as to obtain an upper limit of soot pollution. Eight jars of 800-mL volume were pushed vertically into the top of a sastruga; then a trench was dug along the row of jars so that they could be pulled out sideways. The meltwater from four jars (or eight jars for the cleanest samples) was passed through one filter to make one sample (water volume typically 1 or 2 L). Because soot can collect on the walls of the glass jar and the plastic funnel attached to the filter [Ogren *et al.*, 1983], it is important to minimize the time that the meltwater is in contact with these surfaces. We were able to reduce the time to about 4 min by melting the snow in a microwave oven. Duplicates of a few snow samples (including the remote background samples described below) were shipped frozen to Seattle, where we reduced the time between melting and filtration to 1 min.

Filters were analyzed for their content of light-absorbing impurities by the integrating-sandwich (IS) method [Clarke, 1982]. Application of this method to particles extracted from

snow meltwater using nuclepore filters has been discussed by Clarke and Noone [1985]. Air samples collected on quartz fiber filters (Pallflex, QAS2500) were also measured using the IS technique and calibrated following procedures presented elsewhere [Clarke *et al.*, 1987]. In all cases the change in transmission of light through the IS at a given wavelength can be interpreted as an equivalent absorption by a certain mass of absorbing material per unit area of the filter. The resulting concentration of absorbing material can then be directly expressed as a concentration in mass per unit mass of meltwater extracted (e.g., ng/g).

The two most common light-absorbing particles in the atmosphere are soot and crustal dust. Under most conditions the former dominates aerosol light absorption due to the presence of elemental carbon (EC) that is a major component of soot. EC has a mass absorption coefficient, B , that is several orders of magnitude greater than most crustal dust at visible wavelengths. We have used a value of $B = 9.68 \text{ m}^2/\text{g}$ which Roessler and Faxvog [1980] found for our calibration aerosol (Monarch 71, Cabot Corp.) at 550 nm and which also implied optical properties consistent with our measurement techniques [Clarke *et al.*, 1987]. This value of B will be used here in order to infer an equivalent EC concentration from our absorption measurement. However, it should be kept in mind that this value depends on various properties of the aerosol [Clarke *et al.*, 1987] and can be expected to range from about 5 to 10 m^2/g in the atmosphere. By comparison, we have measured B for our reference dust (clean El Chichon volcanic dustfall with particle diameter $< 1.5 \mu\text{m}$) to be $\sim 0.02 \text{ m}^2/\text{g}$ [Clarke *et al.*, 1983].

Apart from the difference in B for the two light-absorbers, the wavelength dependence of absorption is much steeper for crustal dust than for soot and can be used to infer the

relative contribution of either to the total absorption [Clarke and Noone, 1985]. We have chosen an operational definition of this dependence such that

$$\text{slope} \equiv \frac{(\Delta I/I_0)_{640} - (\Delta I/I_0)_{540}}{0.640 - 0.540} \mu\text{m}^{-1} \quad (1)$$

where I_0 is the intensity incident on the filter and ΔI is the change in intensity after passing through the filter. For the data shown here the slope was determined for the difference in absorption either between 540 and 640 nm or between 525 and 660 nm wavelength.

EXTENT OF SOOT POLLUTION IN SNOW NEAR THE STATION

Figure 1 shows the sites of snow sampling and contour lines based on the concentration of EC found at each site. Each value is the average of at least two samples filtered at the station. Instrument precision for repeated measurements on a single sample was equivalent to less than ± 0.01 ng/g, and thus the uncertainty of soot content is largely due to other causes. The average absolute deviation of individual replicate samples from the mean at particular sites was 0.06 ng/g. At a few selected sites extra samples were taken and stored frozen, to be melted and filtered at the University of Washington. The filters from these samples agreed well with the samples melted at the south pole (average deviation 0.05 ng/g).

The average slope of the wavelength-dependence of absorption was $-0.65 (\pm 0.25) \mu\text{m}^{-1}$, i.e., even less steep than that expected for pure EC (slope ≈ -0.81), indicating that crustal dust cannot be a significant contributor to the observed absorption.

Figure 1 shows that most of the EC is deposited in the prevailing downwind direction. The pattern indicates that the major source of soot is the station, not the runway. However, the airplanes are on the runway only briefly but then spend from half an hour to two hours parked next to the station with engines idling, so that aircraft exhaust could be responsible for a substantial fraction of the soot.

The highest levels of soot found in a significant area of snow downwind from the station, 3 ng/g, when used in a radiative-transfer model, are calculated to reduce the snow albedo by only 0.002 at the most sensitive wavelength (0.5 μm) if present as an "external mixture" [Warren and Wiscombe, 1985, Figure 2] or as much as 0.004 if present as an "internal mixture" [Chylek et al., 1983; Bohren, 1986]. This is below the limit of detectability (0.005) of the spectrophotometers used by T. C. Grenfell et al. (manuscript in preparation, 1990). The quantitative effect of soot on snow albedo was not examined experimentally at the south pole. However, the radiative-transfer model has been validated for larger soot amounts where the effect on albedo becomes detectable [Grenfell et al., 1981; Warren, 1982].

Upwind of the station, where all radiation measurements for climate monitoring have been made, the EC concentration is far too small to affect snow albedo measurements. Indeed, the spectral albedo measurements agreed well with model calculations for pure snow (T. C. Grenfell et al., manuscript in preparation, 1990).

SOOT IN REMOTE ANTARCTIC SNOW

To find a background level of EC in antarctic snow, we collected samples at a site 13 km upwind of the south pole

station (in the 040° direction), while an automatic weather station was being installed. The wind direction for the previous few days had been from 000°. Snow samples were collected from new drift snow, from a soft ridge, and from a sastruga, all about 200 m upwind of the vehicle used to reach the site. The EC concentration was 0.3 ng/g in the ridge and sastruga, and 0.1 ng/g in the new drift snow (Table 1a). (The eleven sites close to the station outside the 0.5 ng/g contour in Figure 1 have an average EC concentration of 0.31 ng/g, indicating that background levels can be found without going great distances from the station.)

These values are about a factor of 10 smaller than that recently reported as an antarctic background value by Chylek et al. [1987]. They analyzed only one sample, which was taken at 48 cm depth from Siple Dome above the Ross Ice Shelf, which is closer to the coast than the south pole is (440 versus 1300 km) and probably has a higher snow accumulation rate. It is not obvious that these differences between the sites could be responsible for the tenfold difference in EC concentrations. We cannot rule out the possibility that part of the difference is due to different sampling and analysis techniques. For example, Chylek et al.'s blank samples gave EC estimates of 0.6–0.9 ng/g, so they would be incapable of measuring values as low as those we report in Table 1a.

CONTRIBUTION OF SOIL DUST TO THE OBSERVED ABSORPTION

The wavelength dependence of absorption of light by aerosol on the filters indicated that the absorption of visible light in South Polar snow was almost entirely due to EC, with little contribution from soil dust or volcanic ash. This is consistent with the results for air filters of Hansen et al. [1988], who were able to attribute essentially all the absorption in south polar air to EC.

The possible contribution of dust to the total absorption in snow remote from the station may also be estimated by measurements of dust content in ice cores. The total number-concentration of particles with diameters greater than 0.63 μm was reported for firn and ice at the south pole by Mosley-Thompson and Thompson [1982], but for estimates of radiation absorption we need the complete size distribution. The total mass concentration of dust in snow has been measured as 15 ng/g at south pole [Kumai, 1976] and 26 ng/g at Dome C [Royer et al., 1983], where the snow accumulation rate is less than half that at the south pole.

Royer et al. used light-scattering measurements on the dust to estimate an effective imaginary refractive index $k = 0.005 \pm 0.005$. As Bohren and Hoffman [1983] have emphasized, no pure minerals have $k \sim 10^{-2}$ in the visible. A value of $k = 0.005$ is an "effective" value for an inhomogeneous dust consisting mostly of transparent minerals ($k \ll 10^{-3}$) but containing a small amount of strongly absorbing material ($k \sim 1$). Using a single effective value of k in computations of radiative transfer through an inhomogeneous dust cannot correctly predict both the absorption and the scattering pattern, but it is all that is available to us and should give a reasonable estimate of absorption. Royer et al.'s estimate agrees with those of other workers for soil dust. Patterson et al. [1977] found k to increase from 0.004 (at 700 nm) to 0.02 (at 300 nm) for Saharan dust, whereas Lindberg and Laude [1974] obtained 0.006 (red) to 0.010 (blue) for desert dust in New Mexico. Egan and Hilgeman [1979] reported k to range

TABLE 1a. Soot in Remote Antarctic Snow

Site Description	Where Filtered ^a	Volume of Meltwater, mL	Soot on Filter, ^b $\mu\text{g}/\text{cm}^2$	Estimated Soot in Snow, ng/g	Slope ^c
New drift snow	SPO	950	0.027	0.10	-0.24
	SEA	1812	0.096	0.10	-0.66
Soft sastruga	SEA	910	0.135	0.28	-0.73
	SEA	1172	0.175	0.28	-0.89
Soft ridge	SPO	1057	0.073	0.25	-0.50
	SEA	1205	0.158	0.24	-0.77
	SEA	878	0.157	0.34	-0.73

Samples were collected 13 km upwind of the South Pole Station, in the 040° direction.

^aSPO means melted and filtered at South Pole Station (filter area 3.64 cm²); SEA means shipped frozen to Seattle and melted and filtered there (filter area 1.89 cm²).

^bAssuming mass absorption coefficient $B = 9.68 \text{ m}^2/\text{g}$.

^cA slope of ≥ -0.8 indicates that all absorption is due to elemental carbon (equation (1)).

from 0.0003 to 0.007 in the visible for 22 common minerals.

To obtain the mass absorption coefficient B from k using Mie theory requires knowledge of the size distribution. The size distribution of dust was not measured at the south pole, but Royer et al. measured it at Dome C using two different methods, finding approximately a log-normal distribution with number-mode radius 0.25–0.5 μm and a mass-mode radius about 1 μm . Most of the dust particles are therefore small enough that they are “volume-absorbers,” and a simple relationship between B and k is approximately valid: $B = 4\pi k/\lambda\rho$. Using $\rho = 2.5 \text{ g}/\text{cm}^3$ for the density of dust, at a wavelength $\lambda = 0.54 \mu\text{m}$, and $k = 0.005 \pm 0.005$, we obtain $B = 0.0465 \pm 0.0465 \text{ m}^2/\text{g}$. This is about a factor of 200 smaller than B for soot given above. Thus a dust concentration of 15 ng/g would cause the same absorption as a soot concentration of $0.07 \pm 0.07 \text{ ng}/\text{g}$. Since we estimated 0.1–0.3 ng/g soot in the remote snow, this implies that about 25% \pm 25% of the absorption would be due to dust. The fact that the slope of absorption versus wavelength in our samples gave no indication of a significant contribution by dust suggests either that k is much smaller than 0.005 for the dust that reaches Antarctica, or that it is less red and more gray in color than either Saharan dust [Patterson et al., 1977] or El Chichon volcanic ash. Further study on this topic would be desirable.

SOOT IN THE REMOTE ANTARCTIC ATMOSPHERE

Since soot incorporated into snow must originate in the atmosphere it is expected that a relationship exists between concentration in the air and in the snowfall. This relationship is frequently expressed as a scavenging ratio defined as the

grams of EC per gram of snow divided by the grams of EC per gram of air. In an earlier paper [Clarke and Noone, 1985] we argued that our snow and air data for the Arctic suggested a regional scavenging ratio of about 160, a value within the range recently determined for submicrometer aerosol removed to the Greenland snowpack [Davidson et al., 1985]. More recently [Noone and Clarke, 1988], we found values between about 60 and 160 for simultaneous measurements in Arctic air and snowfall for a wide range of atmospheric concentrations.

Atmospheric EC concentrations were sampled four times at the South Pole during the snow sample collection period. Samples of ambient air were passed through quartz fiber filters for analysis of EC. We made use of one of several ports on the aerosol sampling stack at the Clean Air Facility (CAF). The intake for this stack, described by Komhyr [1983], is on the roof of the CAF and was about 8 m above the snow surface at the time of sampling (January–February 1986). The volume of air sampled was measured by a flow meter (Dwyer Rotometer) and corrected to the station pressure (680 mbar) and temperature at the flow meter (20°C). The equivalent flow of ambient air at the stack inlet temperature of -35°C was 24.8 L/min. The resulting volumes of air sampled are given in Table 1b.

The air flow had to be interrupted many times to avoid taking air that was contaminated by aircraft exhaust because while airplanes were taking off the wind often was coming across the runway to the CAF. The air flow also had to be interrupted many times to avoid sampling the exhaust from vehicles driving near the CAF. The filters were analyzed by the IS method; the results for all four filters are shown in

TABLE 1b. Air Samples Taken at South Pole, 1986

Filter Elapsed Time		Run Time, hours	Volume, m ³ at -35°C	Soot on Filter, ^a $\mu\text{g}/\text{cm}^2$	Estimated Soot in Air, ^b ng/m ³ or ng/kg	Comments
Start	Stop					
Jan. 25	Jan. 27	45.2	67.3	0.028	1.51	clean
Jan. 27	Feb. 1	117.4	174.5	0.110	2.29	contaminated
Feb. 1	Feb. 6	116.2	172.1	0.077	1.63	contaminated
Feb. 6	Feb. 14	140.0	208.3	0.076	1.32	clean

^aAssuming mass absorption coefficient $B = 9.68 \text{ m}^2/\text{g}$.

^bThe density of air at -35°C and 680 mbar is 0.97 kg/m³ so the units given are essentially interchangeable.

Table 1b. The two "clean" periods suggested atmospheric EC concentrations of about 1.4 ng/m^3 , while those with possible contamination were slightly higher. Contaminated samples are included in Table 1b to illustrate the danger of unattended sample collection. The second filter sampled air continuously for 5 days (January 27–February 1), during which time there were seven airplane takeoffs, some directly upwind of the CAF, and five incidents of snow tractors near the CAF. Some of these events caused spikes on the continuously recording CO_2 gas analyzer at the CAF. These brief events, representing less than 1% of the sample time, are apparently responsible for causing the EC content of the filter to be nearly double that of the "clean" samples. Air sampling on the third filter was terminated after 5 days because of a pollution event caused by an unannounced trip by a vehicle into the clean-air sector.

An independent continuous monitoring program for EC was initiated at the south pole the following austral summer and was recently reported to yield soot concentrations varying with season from 5 ng/m^3 in December 1986 to 0.5 ng/m^3 in May 1987, with an average of about 1.2 ng/m^3 for January–February 1987 [Hansen et al., 1988]. The close agreement with our result, using a different technique, suggests that our two "clean" samples may be considered representative.

Using an EC concentration in "clean air" of 1.4 ng/m^3 and a concentration in snow of 0.2 ng/g we obtain a scavenging ratio for the south pole in January of about $150 (\pm 50)$. This is based on a density of air of 0.97 kg/m^3 at 680 mb and -35°C . The remarkable consistency of the scavenging ratio for the Arctic and Antarctic snowfall suggests that a common removal mechanism may be involved. It also suggests that the application of this ratio to known snowfall and measured atmospheric EC concentrations could yield a simple means of estimating soot removal and deposition for diverse remote regions. However, the agreement could be accidental; we do not know that the EC removal mechanisms are the same in the Arctic and Antarctic.

CONCLUSIONS

Soot concentrations measured in snow around the South Pole Station are too small to affect snow albedo measurably at that site. However, soot concentrations are elevated and describe a "plume" in a direction downwind from the station, with surface concentration that can be an order of magnitude higher than clean "background" levels upwind of the station.

Concentrations in snow upwind and distant from the station were about 0.2 ng/g while atmospheric concentrations were about 1.4 ng/m^3 of air. The resulting scavenging ratio of soot in the Antarctic, about 150, is in good agreement with values recently established in Arctic regions.

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