Environmental research

Measurement of combustion effluent aerosols from Amundsen–Scott South Pole Station

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Project summary

T his project is part of the Antarctic Environmental Research Program, helping to assess the possible environmental impacts of human activities in Antarctica on the pristine surroundings. One of these impacts is due to the emission of "black carbon" (BC) particles in the combustion exhaust from diesel-powered generators, oil-fired space heaters, and vehicles used to support antarctic operations. When deposited to the snow and ice, BC has a number of environmental effects:

- It may be preserved indefinitely.
- Its high optical absorption will alter the surface albedo and optical properties of the ice.
- Its catalytic activity may lead to modifications of snow and ice chemistry.
- Its associated fuel-derived organic species such as polycyclic aromatic hydrocarbons and other toxics may have a deleterious effect on marginal biota near coastal stations.

Previous work showed that extremely small, but nonetheless detectable, concentrations of BC aerosol are brought to the South Pole in the "background" atmosphere, consistent with models of long-range atmospheric circulation. A 1986 study of the soot concentrations in surface snow and ice at the South Pole showed a clear plume of deposition downwind of the station. In this project, we installed equipment upwind and downwind of the Amundsen–Scott South Pole Station to reestablish the background aerosol measurements and to monitor the station's combustion emissions.

Project activities in 1997

I deployed to Antarctica (I was at South Pole from 1 to 10 February 1997) and installed two Aethalometer[™] instruments at the South Pole Station to measure the concentration of optically absorbing aerosol BC particles suspended in the air. The instruments are automatic and require no operator attention other than periodic checking. Approximately every 2 weeks, the data files from each instrument are transmitted by e-mail, assembled into spreadsheets, and merged with the station's meteorology data.

The upwind instrument at the Atmospheric Research Observatory (ARO) records data on a timebase of 1 hour. Very low concentrations of BC are measured, representing the



Figure 1. Smoothed time-series BC data from instrument at upwind location in ARO building. Extremely low concentrations of BC are measured in the background atmosphere in austral winter. The occasional large spikes are either from station emissions (under conditions of reversed prevailing winds) or local contamination.

transport of polluted air to the pole from lower latitudes. Figure 1 shows the smoothed time series of data from this upwind location. Mean aerosol BC concentrations decreased from 3 nanograms per cubic meter (ng/m³) at the end of austral summer to less than 0.1 ng/m³ in the middle of winter. The reduction is due to the formation of the polar vortex, which prevents incursions of polluted air from lower latitudes. Occasional events of elevated BC occur from time to time, due either to a shift in prevailing wind direction so that the station's plume blows toward the ARO site or perhaps to vehicle operation near the building.

The downwind instrument at the Balloon Inflation Facility (BIF) records data every 10 minutes. The BIF is the farthest away downwind building that is heated and powered during the winter season: the sampling inlet is approximately 270 meters from the main exhaust stacks of the central generating plant. High concentrations of BC are measured whenever the wind blows the exhaust plume over the sampling location. Figure 2 shows a typical 2-week record from the downwind



Figure 2. Ten-minute time-series data from instrument at downwind location in BIF building. BC concentrations are very low when background air is sampled; when the wind vector sweeps exhaust emissions over the sampling inlet, the BC concentrations increase by 5 or 6 orders of magnitude.

instrument. When the wind is not *directly* from the exhaust stacks, the BC concentrations are low. Whenever the exhaust plume passes over the measurement point, we see giant excursions in the BC data.

We merged the wind speed and direction data from the meteorology files with our BC concentration results. The emission source strength—i.e., the number of grams of BC emitted per second from the diesel engines—is expected to be approximately constant, although the exhaust is dispersed into a wind of varying speed. Consequently, rather than examining BC concentration versus wind direction, we calculate the flux



Figure 3. Flux of BC (concentration times wind speed) passing over the downwind BIF location, gathered into 10-degree sectors of wind direction. The solid line shows the BC flux data; the broken line shows the number of 10-minute data points included in each 10-degree wind direction sector.

of BC passing the measurement point by multiplying the concentration and the wind speed.

Figure 3 shows the BC flux data gathered into 10-degree sectors. The vast majority of data points lie in the vector range of $+90^{\circ}$ to -45° , representing the prevailing wind direction. Individual flux measurements up to almost 350 micrograms per square meter per second ($\mu g/m^2/sec$) are calculated when the wind direction lies in the sector from $+10^{\circ}$ to $+30^{\circ}$, which corresponds to the +22° grid angle from the exhaust stack to the BIF building. The solid line shows the sector-averaged BC flux, peaking at about 20 μ g/m²/sec for the 10–20° sector. The broken line shows the number of data points represented in each sector and clearly shows the dominance of the prevailing wind. Occasional high flux values are recorded for wind directions up to +90°. Very few data points are seen with wind directions outside this dominant range, and none of those show high BC values. These results confirm our expectation of the impact of the exhaust plume from the generator stack passing the measurement point.

We had earlier estimated that the emission rate of aerosol BC from the main diesel generators at the South Pole Station is on the order of 1,000 µg/sec, based on fuel consumption rates and typical emission indices. The highest 10-minute average flux computed from the downwind instrument's data is over $300 \mu g/m^2/sec$ and is consistent with the emission estimate.

Application of results to an assessment of environmental impact

T he data suggest that the emission rate of aerosol BC from the main generators of the South Pole Station is indeed on the order of 1 milligram per second (mg/sec). The upwind data show that the BC concentration in the background atmosphere is in the range of 0.1 to 0.3 ng/m³ for several months. Because the background air is so clean, the emissions will be detectable in a plume expanding to a width of some tens of kilometers, extending some hundreds of kilometers downwind. The plume of combustion exhaust particles from the South Pole Station would sweep around with the wind direction and be detectable over background—i.e., represent a measurable environmental impact—at great distances from the station itself.

This impact represents only that from year-round, winter season activities—i.e., the steady emission of soot from the main generators. It is my personal belief that summer season activities are likely to lead to *far greater* emissions of soot from aircraft operations, intensive vehicle operations, heating of outlying buildings, and so forth. As this project proceeds, our ongoing data collection will contribute to the assessment of the environmental impact from combustion emissions at the South Pole Station.

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PM₁₀ source apportionment at McMurdo Station, Antarctica

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he National Science Foundation (NSF) Environmental Research Program "support(s) research that can help reduce the environmental footprint of NSF's activities in Antarctica and establish a baseline for future measurements." PM_{10} (atmospheric aerosol particles with diameters less than 10 microns) is a regulated air pollutant in the United States. A study was undertaken to determine the natural and anthropogenic sources of PM₁₀ at McMurdo Station (77°51'S 166°41'E), whose austral summer population exceeds 1,000 and whose reported summer diesel fuel consumption (Antarctic Support Associates 1994) for power generation, heating and water production, and equipment operation is 6.1 million liters. An understanding of the nature and sources of atmospheric particulates at McMurdo Station will provide a basis for evaluating their effects on the antarctic environment.

Air-sampling was initiated at McMurdo Station during austral summer 1995–1996. Emissions were sampled from the major sources: power generators, space heating, surface vehicles, helicopters, and C-130 aircraft. Local soils from around the station were also collected. PM_{10} monitoring was begun at Hut Point, located 600 meters (m) northwest and downwind of McMurdo center. Radar Sat, located on a ridge northeast of the station, was used as a background site. Forty-eight-hour–duration PM_{10} samples were collected continuously at both sites from November through January during the austral summers of 1995–1996 and 1996–1997.

Average PM_{10} concentrations (μ g/m ³) at Hut Point and
Radar Sat during austral summer 1995–1996

	Hut Point		Radar Sat	
Species	Ships (N=6)	No ships (N=27)	(N=32)	
Mass	5.3±2.9 ^a	3.2±1.2	4.8±4.0	
Organic carbon Elemental	0.64±0.50	0.146±0.097	0.17±0.36	
carbon	1.50±2.12	0.171±0.091	0.055±0.034	
Sulfate	0.69±0.29	0.54±0.17	0.69±0.31	
Nitrate	0.034±0.009	0.051±0.020	0.049±0.023	
Ammonium	0.073±0.013	0.066±0.021	0.081±0.048	
Sodium	0.168±0.060	0.24±0.16	0.26±0.22	
Silicon	0.45±0.11	0.41±0.26	0.84±0.85	
MSA ^b	0.132±0.129	0.112±0.108	0.106±0.106	
^a Average ± standard deviation ^b Methanesulfonate				

Source and ambient samples were analyzed for particle mass, ion, element, and elemental and organic carbon (EC and OC) concentrations. Source profiles (fractional abundances of each chemical component with respect to emitted mass) were determined for the major sources. Source apportionment is accomplished using the chemical mass balance (CMB) model (Watson et al. 1990), which compares the source profiles and ambient PM_{10} chemical concentrations. The results are source contributions to PM_{10} mass and its chemical components.

Average concentrations (µg/m³, micrograms per cubic meter of air) of PM₁₀ and selected chemical species at Hut Point and Radar Sat are given in the table for austral summer 1995–1996. Samples affected by emissions from ships docked at the ice pier near Hut Point exhibited much higher EC and OC concentrations than those not so affected. The average EC concentration at Hut Point (no ships) (0.171 µg/m³) was 100-200 times higher than that reported for the South Pole during austral summer (Bodhaine 1995). EC concentrations at Radar Sat were roughly 3 times lower than those at Hut Point, suggesting local contamination, perhaps by motor vehicles working in the area or visiting the site. The higher silicon concentrations (indicative of resuspended geological dust) at Radar Sat may also reflect disturbance of the exposed ground surface by vehicles. Note that average PM₁₀ mass concentrations (excluding ships at Hut Point) were more than a factor of 10 lower than the U.S. Environmental Protection Agency annual average PM_{10} standard of 50 µg/m³.



Figure 1. CMB source apportionment at Hut Point, austral summer 1995–1996. (ug/m³ denotes micrograms per cubic meter.)

Source contributions to PM_{10} at Hut Point were estimated for samples unaffected by ships. The average source contributions are shown in figure 1. Because most nitrate and sulfate in the atmosphere is secondary (formed by gas-to-particle conversion during transport from sources), source profiles representing pure ammonium nitrate, ammonium sulfate, ammonium bisulfate, and sulfuric acid were included in the CMB. Figure 1 indicates that the largest PM_{10} component was geological dust (46 percent). This material probably would not be present to the same extent in the absence of human activity. The next largest source is sea salt (21 percent), originating from nearby McMurdo Sound and the open ocean some 40 kilometers (km) north of McMurdo. Combustion sources accounted for 0.51 µg/m³, or 17 percent.

Ammonium nitrate accounts for only 2 percent of PM_{10} . It is likely, however, that nitrate at McMurdo is associated with large sea salt particles and not ammonium (Mamane and Gottlieb 1992). Nitrate concentrations at McMurdo (table) were similar to levels (0.043 µg/m³) measured at coastal Mawson, Antarctica, during austral summer (Savoie et al. 1992), suggesting that McMurdo nitrogen oxide emissions had not been converted to particle nitrate in the short distance between McMurdo and Hut Point.

Secondary sulfate is a significant component of PM_{10} (15 percent). This amount represents sulfate not associated with geological dust, sea salt, or primary (emitted directly as particles) combustion emissions. We refer to it as "excess" sulfate (xSO₄). It appears to be acidic, e.g., sulfuric acid and ammonium bisulfate. The equivalent concentration of xSO₄ ion is 0.40 µg/m³. This quantity is higher than non-sea salt sulfate (NSS) concentrations (0.21 µg/m³) reported for Mawson during austral summer (Savoie et al. 1992) but lower than summer NSS concentrations (0.65 µg/m³) measured near the northern tip of Ross Island (Wylie et al. 1993). McMurdo sulfur dioxide emissions have apparently not been converted to sulfate en route to Hut Point.

Some sulfate in the marine boundary layer has a biogenic origin. Dimethylsulfide emitted by phytoplankton oxidizes to sulfate and methanesulfonate (MSA). The ratio of MSA to NSS shows a latitudinal dependence that is related to air temperature (Bates, Calhoun, and Quinn 1992). It has been hypothesized that variations in the MSA/NSS ratio in antarctic snow and ice are related to large-scale climatic and meteorological factors (Legrand and Feniet-Saigne 1991).

Based on the relationship described by Bates et al. (1992) (MSA/NSS (molar ratio) = $-0.015(T^{\circ}C)+0.422$), the expected ratio at McMurdo, with a summer average temperature of $-2.4^{\circ}C$, is 0.46. The average MSA/average xSO_4 molar ratio at Hut Point was 0.28. Summer ratios in aerosol at Mawson (Savoie et al. 1992) and on the east antarctic plateau (De Mora, Wylie, and Dick 1997) were 0.18 and 0.20, respectively. The ratio in east antarctic plateau surface snow was 0.08 (De Mora et al. 1997). Low ratios on the plateau may be related to transport from low latitudes and/or to loss of larger particle MSA during transport.

Excess sulfate at Hut Point is not necessarily biogenic. Figure 2 shows the relationship between MSA and xSO_4 at Hut Point (no ships). The regression equation in figure 2 implies an MSA/biogenic sulfate ratio of 0.56 and a nonbiogenic xSO_4 background of $0.2 \,\mu g/m^3$.

Mount Erebus has been discounted as a significant source of sulfur on the antarctic plateau (Delmas 1992). Mount Ere-



Figure 2. Relationship between MSA and excess sulfate at Hut Point. (ug/m³ denotes micrograms per cubic meter.)



Figure 3. Sulfate sources at Hut Point. (ug/m³ denotes micrograms per cubic meter.)

bus may be a significant source of sulfur in the Ross Island area, where down-slope flow from the volcano may enhance surface sulfate aerosol concentrations and may explain why NSS and xSO_4 concentrations are higher on Ross Island than at Mawson. Figure 3 presents a sulfate source budget for Hut Point derived from the above analyses. Sea salt and resuspended geological dust together account for 25 percent. Combustion and biogenic emissions account for 13 and 37 percent, respectively. The remaining "other" 37 percent may represent a hemispheric background and/or emissions from Mount Erebus. It is clear from the elevated elemental carbon concentrations at Hut Point that McMurdo has at least perturbed the atmosphere around the station. The potential impact of associated air pollutants such as polycyclic aromatic hydrocarbons and oxides of nitrogen and sulfur on downwind ecosystems is of great interest. As part of this study, meteorological modeling will be used to estimate the areal extent of the McMurdo plume.

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Clean-air monitoring at Amundsen–Scott South Pole Station, 1996–1997

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he National Oceanic and Atmospheric Administration (NOAA) has had a continuous presence at the South Pole since 1971. NOAA's Climate Monitoring and Diagnostics Laboratory (CMDL), based in Boulder, Colorado, has established and now maintains four baseline observatories in remote locations around the world: Point Barrow, Alaska; Mauna Loa, Hawaii; Cape Matatula, American Samoa; and Amundsen-Scott South Pole Station, Antarctica. CMDL conducts research related to atmospheric constituents that are capable of forcing change in the climate of Earth through modification of the atmospheric radiative environment, and those that may cause depletion of the global ozone layer. CMDL accomplishes this goal primarily through long-term measurements of key atmospheric species such as carbon dioxide (CO₂), carbon monoxide, methane, nitrous oxide, surface and stratospheric ozone, halogenated compounds including CFC replacements, aerosols, and solar and infrared radiation at the baseline observatories and through cooperative measurements at other sites spanning the globe. Through these measurements, CMDL documents global changes in those species important for climate forcing and depletion of the ozone layer, identifying sources, sinks, and interannual variability. Mark Boland, LT, NOAA Corps (Officer-In-Charge) and Glen McConville (Electronics Technician) operated the CMDL program at South Pole during the 1996–1997 season.

Special conditions at the South Pole make it an ideal location for monitoring the composition and behavior of the atmosphere.

- Local contamination is minimal, as are regional-scale sources and sinks. The nearest city is 4,800 kilometers away, and the nearest land biota is 4,000 kilometers away.
- Because contamination sources are distant, the air measured at the South Pole is very well mixed and is extremely clean. When compared to measurements made elsewhere around the globe, it is truly the background air of the planet.

- Over 96 percent of the time, the wind blows from the Clean Air Sector at the South Pole. Data capture is very high, and collection of a long-term continuous record is greatly facilitated. It is easy to "flag" the few periods when air comes from the station.
- The water vapor concentration in the atmosphere is extremely low.
- The South Pole will remain an excellent baseline site, because upwind areas will not be developed.
- In combination, the antarctic continent, surrounding ocean, and overlying atmosphere act as a controlling governor (a heat sink) for the world's climate engine, and measurements at the South Pole are expected to provide early warning signals for identifying atmospheric composition and surface-energy budget changes that will lead to new climate and weather patterns on global and regional scales.

A brief summary of the CMDL program during the 1996–1997 season follows.

Greenhouse gases

C arbon dioxide is measured continuously using a nondispersive infrared analyzer. Atmospheric CO_2 concentrations continue to increase globally. The long-term record from South Pole, presented in figure 1, shows a growth rate of approximately 1.4 parts per million per year (Hofmann, Peterson, and Rosson 1996).



Figure 1. Monthly average of CO_2 mixing ratios in parts per million (ppm) from *in situ* measurements.

Halocarbons

We are using two electron-capture gas chromatographs to make continuous measurements of various halocompounds, and as a complement to this, steel flasks are pressurized with outside ambient air and are then sent to Boulder for analysis.

Our research has shown that the rate of growth of chlorofluorocarbons (CFCs) is decreasing and that their concentrations should reach maximum levels in the stratosphere within the next several years (see figure 2). It is anticipated that the ozone layer will then begin a slow recovery, which, barring any noncompliance with the Montreal Protocol, will be readily detectable by the year 2050 (Hofmann 1996).

Surface and stratospheric ozone

S urface ozone is measured using the Dasibi ozone monitor. Stratospheric ozone is measured using two different instruments. The Dobson spectrophotometer measures total column ozone, and high-altitude balloons are used to determine the atmospheric ozone profile. Ozone measurements at the South Pole go back to 1961 when the first Dobson spectrophotometer measurements were made and, thus, provide an ongoing 35-year record.

Figure 3 shows the ozonesonde profile from the 8 October 1997 flight as compared to a preantarctic-ozone-hole profile from 7 August 1997. This profile clearly shows the near total destruction of ozone in the 14- to 20-kilometer altitude region of the stratosphere. This depletion is typical of the losses seen during the springtime over Antarctica, when nearly two-thirds of the ozone is lost.



Figure 2. Monthly average of trichlorofluoromethane (CFC-11) mixing ratios in parts per trillion (ppt) from *in situ* measurements.

Solar radiation

W e have 10 separate instruments for measuring solar and thermal radiation. They include pyrheliometers, pyranometers, and pyrgeometers.

Aerosols

T hree different instruments are measuring aerosol particles and concentrations.

The nephelometer measures the scattering part of the extinction coefficient due to particles. First, air is pulled into the instrument through a filter that removes all the aerosol particles. Scattered light intensity is measured and recorded as the background. Then ambient air is introduced into the instrument, and the intensity of the scattered light is measured again. Simple subtraction then yields the contribution due to aerosols. This cycle is performed at four wavelengths of light (450, 550, 700, and 850 nanometers) to give a size distribution. Particle sizes measured range from 0.1 to 1.0 micrometers.



Figure 3. NOAA/CMDL ozonesondes profiles of ozone partial pressure in millipascals (mPa) at South Pole during the ozone hole (8 October) and the predepletion profile (7 August).

The Pollack Condensation Nuclei Counter uses light attenuation by cloud formation due to particle growth [from adiabatic expansion of condensation nuclei (CN)] in supersaturated air to measure CN. The CN are typically 0.001 to 0.1 micrometers in radius. These particles are also called "Aitken nuclei."

The TSI CNC also measures Aitken nuclei. It uses a laser diode light source and a single-particle counting optical detector to count individual particles approximately 0.014 micrometers and larger that have been grown onto alcohol droplets.

Meteorology

B ordering the Clean Air Sector is our 23-meter-high meteorological walk-up tower. Located on the tower, we have an anemometer, for determining wind speed and direction; three aspirated and one nonaspirated thermometers; and a hygrometer, for measuring frost-point. Located inside the Clean Air Facility are two barometers.

Data from all of the instruments are transferred electronically to the CMDL offices in Boulder. This data transfer is performed daily, weekly, or monthly depending on the instrument. Most of the instrument data-acquisition systems have programs that automatically "zip" the data files, which can then be FTPed to Boulder for postprocessing and analysis.

In addition to the CMDL-sponsored research, six cooperative research projects are conducted at the South Pole for various universities and other government agencies. These projects include isotopic composition for atmospheric CO₂, CO₂ flask sampling, oxygen and nitrogen flask sampling, snow sampling for hydrogen peroxide, measuring the distribution of specific and anthropogenic radionuclides in surface air (filter sampling), and quantifying the production rate of radiocarbon by galactic cosmic rays.

This program has been operated in cooperation with, and through support from, the National Science Foundation for more than 25 years.

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