

## A controlled snowmaking experiment testing the relation between black carbon content and reduction of snow albedo

Richard E. Brandt,<sup>1</sup> Stephen G. Warren,<sup>1</sup> and Antony D. Clarke<sup>2</sup>

Received 14 November 2010; revised 9 February 2011; accepted 15 February 2011; published 20 April 2011.

[1] Radiative transfer modeling of the reduction of snow albedo by black carbon (BC) requires experimental verification. In natural snow the albedo reduction is at most a few percent, and even with accurate measurements, attribution is ambiguous because snow albedo depends on other variables. In this experiment, artificial snowpacks are made by freezing of water droplets produced by a snowmaking machine in an open field, using water with and without added soot, in amounts about 100 times natural background soot levels, so as to obtain a large signal on albedo. The optically effective snow grain size is determined from the measured near-infrared albedo; matching the measured visible albedo then requires addition of BC to the radiative transfer model. The BC content of the artificial snowpacks is measured by filtering the meltwater; the filters are analyzed by a laboratory spectrophotometer as is done for filters from samples of natural snow. The BC content indicated by the filters agrees with that required in the model to match the observed albedo, but significant uncertainties remain, so further experiments are needed.

**Citation:** Brandt, R. E., S. G. Warren, and A. D. Clarke (2011), A controlled snowmaking experiment testing the relation between black carbon content and reduction of snow albedo, *J. Geophys. Res.*, 116, D08109, doi:10.1029/2010JD015330.

### 1. Introduction

[2] Recent concern about the climatic effects of black carbon (BC), both in the atmospheric aerosol and in snow at the Earth's surface, have motivated modeling, fieldwork, and laboratory studies of (1) the optical properties of carbonaceous particles [Bond and Bergstrom, 2006], (2) methods of BC measurement [Slowik et al., 2007; Boparai et al., 2008; Grenfell et al., 2011; Cross et al., 2010], (3) Arctic air pollution [Quinn et al., 2008], (4) measurements of BC concentration in snow [Doherty et al., 2010, and references therein], and (5) modeling the radiative forcing and climatic impact of BC in snow [Hansen and Nazarenko, 2004; Jacobson, 2004; Flanner et al., 2007]. A comprehensive assessment of the role of BC in climate is underway (T. Bond et al., Bounding the role of black carbon in climate, manuscript in preparation, 2011). Although several research groups have used radiative transfer modeling to compute the reduction of snow albedo for a specified BC concentration, there has not been experimental verification of those calculations. Experimental quantification of the link between BC concentration and reduction of snow albedo is the topic of this paper.

[3] Modeling of radiative transfer in pure snow [Wiscombe and Warren, 1980] was validated by spectral albedo measurements on Antarctic snow, where the impurity content

is too small to affect the albedo measurement [Grenfell et al., 1994; Warren and Clarke, 1990]. The dependence of snow albedo on impurity content was first quantified by the radiative transfer modeling of Warren and Wiscombe [1980], which indicated large reductions of albedo at visible and ultraviolet wavelengths for parts-per-million (ppm) amounts of soot. However, in remote snow of the Northern Hemisphere, the levels of soot pollution are much lower, in the parts-per-billion (ppb) range [Clarke and Noone, 1985; Doherty et al., 2010], where the effect on albedo is at most a few percent. A reduction of albedo by 1–2% is significant for climate but is difficult to detect experimentally and even more difficult to attribute to a cause, because snow albedo depends on several other variables, particularly snow grain size [Wiscombe and Warren, 1980]. In our work to quantify the radiative effect of black carbon (BC) in snow, we therefore do not directly measure the albedo reduction. Instead, our procedure is indirect and consists of two steps. (1) We collect snow samples, melt and filter them, and analyze the filters spectrophotometrically for BC concentration [Grenfell et al., 2011]. (2) We use the BC amount from the filter measurement, together with snow grain size, in a radiative transfer model to compute the albedo reduction.

[4] The quantity required for radiative transfer modeling is the absorption coefficient  $k_{abs}$ , in units of  $m^2/(g \text{ snow})$ . This is obtained from the filter measurement as the absorption cross section of particles on the filter, divided by the mass of meltwater passed through the filter. For convenience in relating our results to the predictions of atmospheric transport and deposition models, we convert  $k_{abs}$  to a concentration  $C$  of BC in snow,

$$k_{abs} = B_a C,$$

<sup>1</sup>Department of Atmospheric Sciences, University of Washington, Seattle, Washington, USA.

<sup>2</sup>School of Ocean and Earth Sciences and Technology, University of Hawai'i at Mānoa, Honolulu, Hawaii, USA.

where  $C$  has units (g BC)/(g snow), and  $B_a$  is the mass-absorption cross section (MAC) of BC ( $\text{m}^2/\text{g}$ ). Our filters are calibrated by reference to standard filters with known (weighed) amounts of Monarch 71 soot, whose MAC was determined by optical analysis of a spectrophotometer to be  $B_a \approx 6 \text{ m}^2/\text{g}$  [Clarke and Noone, 1985; Clarke et al., 1987].

[5] Because the computed reduction of snow albedo is model-based, it requires experimental verification. We doubt that direct measurement of albedo reduction will be feasible in nature, because of the vertical variation of both snow grain size and soot content, and because the natural soot content is small (except very close to some sources, where its size distribution is also unrepresentative). Furthermore, deep snow would be required, because the spectral signature of sooty snow is similar to that of thin snow (visible albedo reduced but near-infrared albedo unchanged; compare Figure 13 of Wiscombe and Warren [1980] to Figure 7 of Warren and Wiscombe [1980]). Also, accurate knowledge of the instrument's shadowing correction would be needed, because its value is typically  $\sim 1\%$ , i.e., of similar magnitude to the albedo reduction for typical soot amounts in Northern Hemisphere snow. For example, in the experiment described below, the presence of the instrument and operator near the targeted snow reduced the downward diffuse irradiance on the snow target by 1.7%, but this value is uncertain to perhaps a factor of 2. In natural snow with only  $\sim 10$  ppb soot, the inferred values of  $B_a$  values, with and without applying a shadowing correction, could differ by a factor of 10 or more.

[6] We conclude that what is needed is an artificial snowpack, with uniform grain size and large uniform soot content (ppm not ppb), so as to produce a large signal on albedo. The experiment can be done in a freezer laboratory or outdoors. The experiment we are pursuing is done outdoors. The reasons for choosing this approach are as follows.

[7] 1. Visible radiation penetrates tens of centimeters into snow, so photons emerge horizontally distant from where they entered. In the limited width of a laboratory snowpack, radiation may be absorbed by the walls of the container before it can reemerge from the snow to contribute to the albedo.

[8] 2. Also because of the horizontal transport of photons before reemergence, it is necessary to have uniform illumination over a large area. This is difficult to achieve in the laboratory, but is easily obtained if the source of radiation is the Sun.

[9] 3. In a laboratory experiment only a narrow field of view can be measured, rather than a hemispheric field of view, so a laboratory experiment measures the bidirectional reflectance for particular angles rather than albedo (the integral of reflectance over the hemisphere).

[10] The disadvantage of an outdoor experiment is that one must wait for appropriate weather: low temperature ( $-20^\circ$  to  $-40^\circ\text{C}$ ), calm winds, diffuse incident radiation for the albedo measurement (for reasons explained in section 3), and no snow falling during the experiment.

## 2. Production of Artificial Snowpack

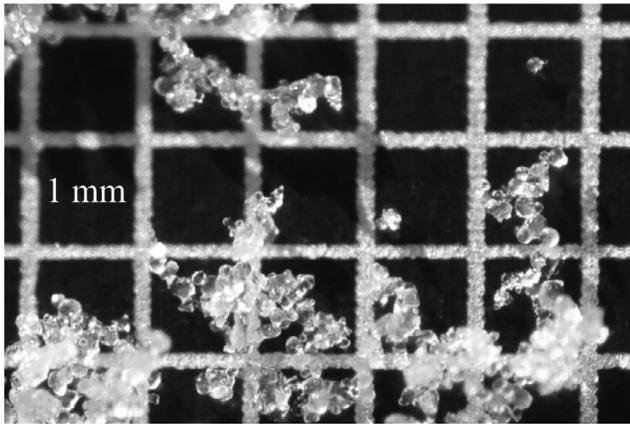
[11] The experiments were carried out on an open field behind the elementary school at Bloomingdale, New York.

A small snowmaking machine, using the village water supply, could make a snowpack of area  $75 \text{ m}^2$  and depth 15 cm in a period of 4 h, deposited over  $\sim 40$  cm of natural snow. (The snow depth was 4 cm liquid equivalent, which is optically semi-infinite according to Figure 13a of Wiscombe and Warren [1980]). The snowmaking machine consisted of a 6-horsepower pressure washer and 4-horsepower air compressor which provide pressurized water and air to a "Snow at Home" brand SG7 Xstream snow gun. The air and water pressure, and the water flow rate, were monitored to ensure stability of the snowmaking process and constancy of the final soot concentration. The compressed air was filtered and desiccated to remove residual oil. A soot suspension in water at about 1000 times the final concentration was maintained in a sonicated bath, which was entrained into the water stream using a Mecomatic injection pump feeding a 4 L mixing chamber to ensure a uniform dilution. Bypass valves enabled switching between the diluted soot suspension in the mixing chamber and pure water.

[12] The snowmaking operation typically began in the evening, about 2200 local time, with a 30 min purge to clean any residue from the system. Two snowpacks were made, first without and then with added soot. The snowpacks were built about 10 m apart and 10 m upwind of the snowmaking machine to avoid cross contamination and contamination from the compressors' exhaust. For a soot content of 1 ppm, 3 g of soot were dispersed into 3 tons of snow. The Bloomingdale water supply contained a small amount of a brown absorber (perhaps rust or humic acid); filter measurements indicated a spectrally integrated absorption equivalent to that of a BC content of  $\sim 10$  ppb ("equivalent BC," as defined by Grenfell et al. [2011]). This water was actually cleaner than newly fallen snow, which contained 20–60 ppb of BC, probably originating from residential wood-burning stoves in the surrounding region.

[13] The artificial snow grains were quasi-spherical, of radii  $\sim 60 \mu\text{m}$  (Figure 1). They were ejected from the nozzle as droplets, which froze in the cold air, so the soot particles were probably uniformly distributed within each droplet, at least for the hydrophilic soots.

[14] Three types of soot were used. The soot that had been used to make the reference standards, Monarch 71, was no longer available; a close match was provided by Monarch 120 from Cabot Corporation. Another commercial soot, Aquablack 162 from Tokai Corporation, has been chemically modified in the manufacturing process to contain some polar groups on the surface so that it is soluble in water, facilitating the experimental procedures. The experiment in which the albedo measurement was most successful used this type of soot. The size distribution of Aquablack (Figure 2) is smaller than that of BC found in ambient atmospheric aerosol and in natural snow, so that its spectral absorption is somewhat stronger at blue wavelengths and weaker at red wavelengths. The small sizes also meant that when the meltwater was passed through our standard  $0.4 \mu\text{m}$  Nuclepore filter much of the soot was not collected, so we passed the filtrate successively through  $0.2 \mu\text{m}$  and  $0.1 \mu\text{m}$  filters. A third soot, "Fullerene" from Alfa Aesar Corp., has also been tried, but so far when we

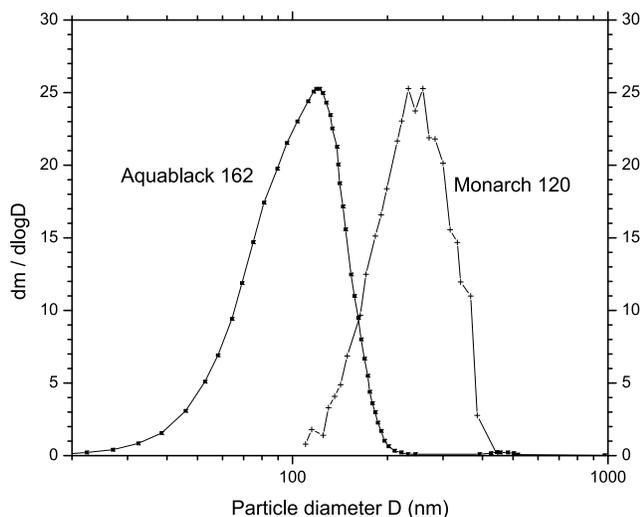


**Figure 1.** Frozen water droplets used to make the artificial snowpack. The grid spacing is 1 mm.

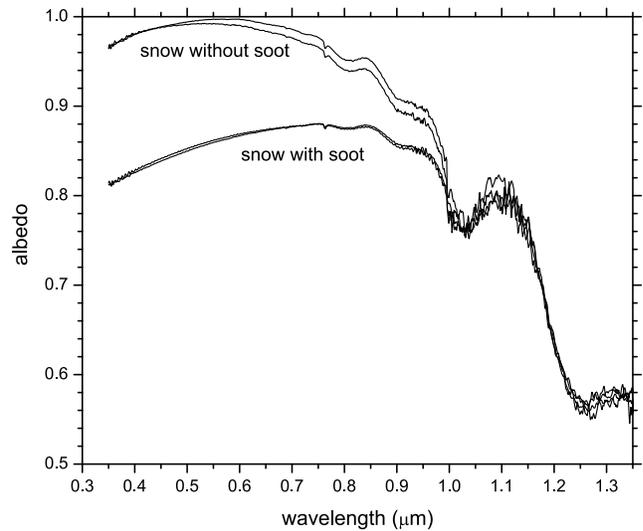
have used it the weather conditions were not suitable for a successful experiment.

### 3. Albedo Measurements

[15] We examine in most detail the experiment of 5 February 2009. Albedo as a function of wavelength was measured with a FieldSpec Pro JR spectral radiometer from Analytical Spectral Devices, Inc. (ASD) [Kindel *et al.*, 2001], equipped with a diffuse reflecting plate to collect radiation with equal efficiency from an entire hemisphere. For highest accuracy, the albedo measurements were made under diffuse incident radiation (overcast sky), thus minimizing errors due to tilt of the instrument, nonhorizontality of the snow surface, and deviation from cosine response of the diffuser plate. The ASD instrument was positioned at two locations on each snowpack; the small differences shown in Figure 3 are probably the result of different contributions from the natural snow surrounding the artificial



**Figure 2.** Size distribution of Aquablack 162, obtained from a differential mobility analyzer (DMA), and of Monarch 120, obtained from a Single Particle Soot Photometer (SP2). The units of  $m$  are mass per unit volume, but scaled arbitrarily.



**Figure 3.** Spectral albedo of two artificial snowpacks on 5 February 2009, one with and one without added soot in the form of Aquablack 162. Measurements from two locations on each snowpack are shown. All values were multiplied by 1.017 before plotting, to account for the shadowing correction of 1.7%. The data are noisier at the longer wavelengths because of weaker incident solar radiation. The analysis of filters through which meltwater was passed indicated a BC content of 12 ppb (of BC equivalent) for the clean snow and 2250 ppb for the sooty snow.

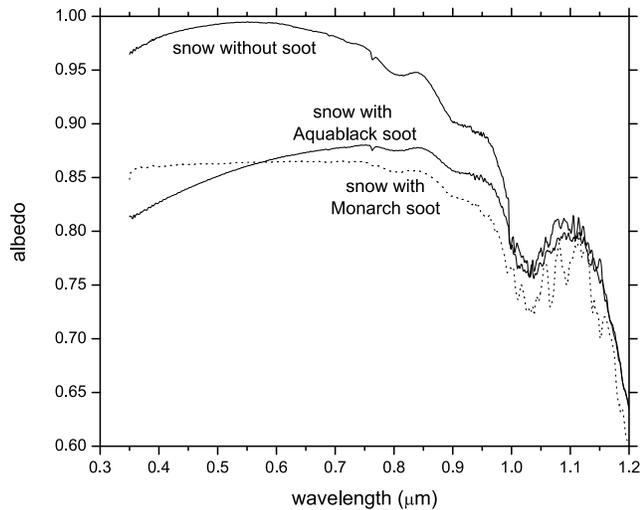
snowpack, whose grain size would have been different. [For the diffuser plate at height 0.7 m over a snowpack of radius 4.9 m, about 2% of the flux received by the radiometer would have originated from outside the artificial snowpack.

[16] Figure 3 shows that the spectral albedos of the dirty and clean snowpacks were in agreement for near-infrared wavelengths  $\lambda > 1.0 \mu\text{m}$ , but diverged at shorter wavelengths. This is as expected, because at near-infrared wavelengths ice itself absorbs significantly, so small amounts of impurities have little effect. The shadowing correction for diffuse incidence was estimated by geometric analysis as 1.7%. The observed albedos were therefore multiplied by 1.017 before further analysis. Figure 3 shows the albedos after the shadowing correction was applied.

[17] Figure 4 compares the experiments with two different types of soot. The smaller sizes of the Aquablack cause its MAC to increase toward shorter wavelengths, resulting in a reddening of the albedo curve, whereas the Monarch 120 results in a reduced albedo that is nearly constant from 0.35 to 0.8  $\mu\text{m}$  wavelength because of its larger particle sizes (Figure 2). The Monarch 120 size distribution approximates that of ambient BC aerosol in Arctic pollution shown in Figure 2 of Schwarz *et al.* [2010]. Figure 4 does not show an albedo measurement for the clean snow in the Monarch experiment, because the wind strengthened during the snow-making operation and blew some of the sooty snow onto the clean snowpack.

### 4. Radiative Transfer Modeling

[18] The sooty snowpack was modeled as an external mixture of soot spheres and ice spheres (Figure 5). The



**Figure 4.** Spectral albedo of three artificial snowpacks. The plots for snow without soot and for snow with Aquablack soot are averages of the corresponding plots for 5 February 2009 in Figure 3. The snow with Monarch soot was measured on 25 January 2009; its filter indicated 870 ppb of BC. At 1.0–1.2  $\mu\text{m}$  the albedo is lower than for the 5 February snow, indicating a larger grain size. The data are also noisier at these wavelengths because of low light levels during the measurement.

near-infrared albedo is sensitive to snow grain size, so the grain size in the model was chosen to match the albedo curves for  $\lambda > 1.0 \mu\text{m}$ , giving an optically effective radius (area-weighted mean radius [Hansen and Travis, 1974]) of  $r \approx 60 \mu\text{m}$  for both the clean and sooty snow, in qualitative agreement with the photograph in Figure 1. The size distribution of the snow grains was modeled as a monodispersion, broadened just enough to average over the interference structure in the Mie extinction efficiency, as had been done by Wiscombe and Warren [1980]. We find that different grain size distributions with the same effective radius produce nearly identical albedos, as was first pointed out for clouds by Hansen and Travis [1974]. The effective radius was determined as the best fit over four narrow bands of width 4 nm centered at 897, 1030, 1090, and 1310 nm.

[19] The soot specifications for the model had been chosen for use in a comprehensive modeling study, to represent ambient soot aerosol in the atmosphere distant from sources. The soot was assigned a density of  $1.8 \text{ g cm}^{-3}$  and a complex refractive index  $m = 1.95 - 0.79i$  [Bond and Bergstrom, 2006]. The soot size distribution was modeled as lognormal, with mass mean diameter 130 nm and log-normal width 1.3. Its mass absorption cross section at  $\lambda = 550 \text{ nm}$  is  $B_a = 6.3 \text{ m}^2/\text{g}$ . The BC content of the model was adjusted until the model matched the observed albedo reduction as a function of wavelength (Figure 5); this required  $C = 2.25 \text{ ppm}$ , after iteration to determine the best fit of both  $r$  and  $C$ . If a shadowing correction had not been applied, the inferred BC concentration in the sooty snowpack would be 10% larger.

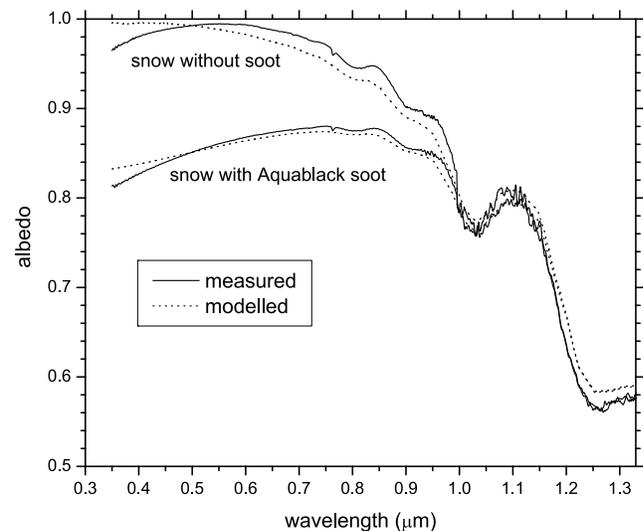
[20] The albedo of the “clean” snow is seen to peak at 0.55–0.60  $\mu\text{m}$  wavelength, decreasing toward the blue and

ultraviolet, in disagreement with the model for pure snow. This is most likely due to the small amount of brown absorber in the water supply. The albedo of snow with soot also decreases toward shorter wavelength, but for a different reason: the particle sizes of Aquablack were somewhat smaller than the size distribution used in the model of atmospheric soot.

## 5. Filter Samples

[21] After completing the albedo measurements, samples of the two snowpacks were collected in glass jars, melted quickly in a microwave oven, and filtered through nuclepore filters, the same procedure used for our samples of natural snow [Doherty et al., 2010]. The filters were processed in an integrating-sandwich spectrophotometer [Grenfell et al., 2011], calibrated by reference to the standard filters containing Monarch 71 soot. The measurement implied 12 ppb for the clean water and 2500 ppb for the sooty snowpack.

[22] This value, 2500 ppb, was obtained from the sum of soot amounts on the sequence of filters:  $4 \mu\text{g cm}^{-2}$  were collected on the 0.4  $\mu\text{m}$  filter,  $2.2 \mu\text{g cm}^{-2}$  on the 0.2  $\mu\text{m}$  filter, and  $1.5 \mu\text{g cm}^{-2}$  on the 0.1  $\mu\text{m}$  filter. The fact that the amount collected on the 0.1  $\mu\text{m}$  filter was only one quarter of the amount collected on the 0.2  $\mu\text{m}$  and 0.4  $\mu\text{m}$  filters, even though essentially all particles of Aquablack are smaller than 0.2  $\mu\text{m}$  (Figure 2), indicates that some clumping of soot particles occurs either in the snowmaking process or in the melting process, or that particles much smaller than the



**Figure 5.** Comparison of measured and calculated spectral albedo for the experiment of 5 February 2009 using Aquablack soot. The snow grain size for the model was chosen to match the near-infrared albedo; it is 57  $\mu\text{m}$  for the clean snow and essentially the same (55  $\mu\text{m}$ ) for the sooty snow. The model for the sooty snow included 2500 ppb of BC as an external mixture. The optical constants of ice were taken from Warren and Brandt [2008]; the BC properties are given in the text. The radiative transfer model used the discrete-ordinates method [Stamnes et al., 1988].

nominal pore size are collected by adhering to the pore walls (we know this from other experiments), or both. Nevertheless, because some soot was collected on the filter with smallest pore size, the actual BC concentration in the snow is probably greater than 2500 ppb, by an unknown amount.

## 6. Discussion and Conclusions

[23] The absorption of light by BC as inferred from the albedo measurements is approximately the same as that inferred from the filter transmission:  $(2.25 \text{ ppm} \times 6.3 \text{ m}^2/\text{g}) \approx (2.5 \text{ ppm} \times 6 \text{ m}^2/\text{g})$ . If this result is substantiated by further experiments, it will mean that filter-derived concentrations (e.g., as reported by Doherty et al. [2010]) can be used directly in a radiative transfer model to calculate albedo reduction. We thus do not require an estimate of the MAC for our routine analyses of natural snow samples; it is enough to know the relation between filter darkness and albedo reduction.

[24] There is however, an important question about the relevance of this experiment to soot in natural snow, namely that the microlocation of soot in natural snow may differ from that in the artificial snow. Although we have no direct evidence, it seems likely that in the artificial snow the soot would be located in the interior of the frozen drops, so its MAC would be enhanced relative to an external mixture by up to a factor of 2 [Ackerman and Toon, 1981; Fuller et al., 1999]. In natural crystals of falling snow, soot can be collected by both nucleation and below-cloud scavenging, so in a natural snowpack the soot particles may be on the surface of snow grains as well as in the interior. Furthermore, postdepositional processes (snow metamorphism) in both dry and wet snow will tend to move impurities from the interior of crystals to the grain boundaries. The albedo reduction we measure for soot in the snowmaking experiment may therefore be greater than the albedo reduction by the same concentration of soot in natural snow, by a factor between 1 and 2.

[25] The experiment with Monarch 120 was not as complete as the Aquablack experiment, but it can be discussed qualitatively. Figure 4 shows that the snow grain size was larger than for the Aquablack experiment, but the snow grain size cannot be quantified because the near-infrared albedo was noisy. However, the larger grain size probably accounts for the fact that the albedo reduction is comparable to the albedo reduction by Aquablack in spite of the smaller BC content as indicated by the filter (0.9 versus 2.2 ppm).

[26] Because Aquablack sizes are smaller than ambient, and because some of the Aquablack may have escaped collection even by the filter of smallest pore size, our validation of the radiative transfer model must be regarded as tentative, awaiting further experiments. A comprehensive study would involve varying (1) the type of soot, to investigate different size distributions; (2) the amount of soot, to investigate the nonlinearity of albedo reduction [Warren and Wiscombe, 1985, Figures 1 and 2], and (3) the snow grain size of the artificial snow, because the albedo reduction by a given concentration of soot is predicted to be greater in coarse-grained snow than in fine-grained snow [Warren and

Wiscombe, 1980, Figure 7]. The grain size may be varied by using different sizes of the nozzle orifice spraying the water into the cold air.

[27] **Acknowledgments.** The principal of Bloomingdale Elementary School, Pat Hogan, kindly made available the necessary utilities and space for the snowmaking experiment. Sarah Doherty, Kristel Guimara, Christina Pedersen, and Ellen Beberman assisted with the experiments. We thank Ross Edwards for recommending the Aquablack soot and Steffen Freitag for measuring the size distribution of Monarch 120. Helpful comments were received from Charlie Zender and two anonymous reviewers. The research was supported by NSF grant ARC-06-12636.

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- R. E. Brandt and S. G. Warren, Department of Atmospheric Sciences, University of Washington, Box 351640, Seattle, WA 98195, USA. (sgw@uw.edu)
- A. D. Clarke, School of Ocean and Earth Sciences and Technology, University of Hawai'i at Mānoa, Honolulu, HI 96822, USA.