

Darkening of Soot-doped Natural Snow: Measurements and Model

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Abstract

We conducted the first measurements of the direct effect of laboratory controlled BC-contamination on snow albedo in a laboratory environment. Optical measurements of natural snow intentionally doped with industrial grade BC were conducted at visible and near-infrared wavelengths. Snow albedo was measured in a (portable) integrating sphere system. The absolute spectral albedos are less than expected at visible wavelengths due to absorption by the sample holder which is too shallow to present a semi-infinite sample surface. Hence we focus on the albedo change by BC doping, rather than on the absolute snow albedo itself. Using preliminary calibrations, our measurements agree well with model predictions that BC concentrations from 1–200 ppmm reduce albedo by 5–70%. A key uncertainty is assessing how much BC mass was “lost” during the doping process and thus did not fully contribute to snow darkening.

1 Introduction

Light-absorbing impurities including black carbon (BC) and dust reduce snow and ice reflectivity (*Warren and Wiscombe, 1980; Chylek et al., 1983*) and can thereby trigger ice-albedo feedbacks (*Warren, 1984, e.g.*). If our representation of the associated processes in climate models is adequate, then snow-darkening by anthropogenic BC deposition is a major contributor to recent Arctic warming (*Hansen and Nazarenko, 2004; Jacobson, 2004; Flanner et al., 2007; Quinn et al., 2008*). Here we report the first laboratory measurements of the direct effect of laboratory controlled BC-contamination on natural snow albedo.

BC darkens snow and alters climate through a series of processes that climate models approximate with varying degrees of uncertainty. The fundamental cause of snow darkening is the contrast between the single scattering albedo of BC, the darkest aerosol, and that of snow, the brightest surface. These single scatter albedos depend, respectively, on the chemical composition, structure, and mixing state of BC (*Bond and Bergstrom, 2005*), and on the size and shape of snow crystals (*Warren and Wiscombe, 1980; Grenfell and Warren, 1999*). Models show that 15 ppbm (parts-per-billion-by-mass) of BC externally mixed in snow of optically-equivalent effective radius $r_e = 100\mu\text{m}$ will darken the snow albedo by about 1% at the mid-visible wavelength $\lambda = 500\text{ nm}$ (*Grenfell et al., 1994*).

Previous measurements of the albedo perturbation of snow by BC have, to our knowledge, tested snow with *a priori* unknown (rather than known) amounts of BC. Typically field snow samples are first collected, melted and filtered. Filters may then be analyzed using thermo-optical

techniques to measure elemental carbon, organic, and/or total carbon mass concentration ([Legrand et al., 2007](#)). A separate approach that requires only optical techniques compares the filter transmission to the transmission observed from known amounts of a “standard” type of commercially available BC ([Noone and Clarke, 1988](#); [Warren and Clarke, 1990](#)). From this one estimates the mass concentration of the standard BC which would have caused the same filter transmission. This method has the advantage of expressing the BC mass in terms of the mass of a well-studied type of BC, facilitating intercomparisons. This optically equivalent BC mass concentration may then be used to estimate the (and/or compare to the field-measured) snow albedo.

Our experiments attempt to perturb the snow albedo with an *a priori* known mass of BC, and then to measure its albedo perturbation. The intent is to test the adequacy and fidelity of forward radiative transfer model treatments of BC under varying conditions, eventually to include other contaminants (e.g., dust) and coatings. To accomplish this we dope natural snow with pre-measured quantities of BC and observe the resulting albedo perturbation.

2 Methods

Our procedure measure the perturbation to snow albedo by BC follows these steps. 1. Collect fresh surface snow. 2. Weigh BC 3. Weigh snow 4. Separately blend control and BC-doped snow samples 5. Dilute snow samples as necessary 6. Measure snow density 7. Measure 1310 nm-reflectance 8. Measure 630 nm-reflectance

We collected surface snow for these experiments during short expeditions to nearby Chamrousse Ski Station (elevation ~ 1750 m). To minimize accumulation of impurities, all specimen collection took place within 24-hours of snowfalls lasting at least 24-hours. Any impurities present in the fresh snow samples contribute to excess absorption in both the clean and doped snow experiments described below.

Prior to collection we measured *in situ* properties (air and snow temperature, snow density and stratigraphy). Near surface air temperature never exceeded $T = -0.5^\circ\text{C}$ between snowfall and collection. Surface specimens (top ~ 50 cm) were transported to LGGE. All subsequent handling, preparation, and measurement of snow took place in a constant-temperature (-15°C) cold-room facility at LGGE.

We doped snow with commercially available Monarch 120 Carbon Black supplied by Cabot Corporation (Billerica, MA, USA). Monarch 120 is most similar to the discontinued product Monarch 71 used to calibrate optically effective BC concentration in many previous studies (e.g., [Clarke and Noone, 1985](#); [Warren and Clarke, 1990](#); [Clarke et al., 2004](#)) (S. Warren, personal communication, 2007). Monarch 120 is not, strictly speaking, soot ([Watson and Valberg, 2001](#)), nor is it pure elemental carbon (EC). Monarch 120 is our experimental analogue for light-absorbing carbon ([Bond and Bergstrom, 2005](#)) aerosol in the climate system, and we refer to it simply as BC henceforth.

BC samples of 3–100 mg were weighed on a precision scale, then mixed into the snow specimens of approximately 440 g. Mixing was accomplished with a commercially available blender composed of aluminum and stainless steel. Blending for one minute, adding BC, then blending for 2.5 minutes produced darkened snow that appeared homogeneous. This mixing procedure created doped snow with initial BC mixing ratios $7.5 \leq q_{\text{BC}} \leq 221$ ppm. Some of this material was diluted with clean snow in the blender to obtain BC mixing ratios as low as $q_{\text{BC}} = 250$ ppb. One

minute of additional blending was performed per dilution stage.

Blank snow specimens were created with the same procedure, but without adding BC. To prevent contamination, separate sets of mixing elements were maintained for the blanks and doped samples. All components were washed in 18 M Ω -cm water between uses, and no residual BC was noticed on the mixing elements.

The key optical measurements were made with DUFISSS, the DUal-Frequency Integrating Sphere for Snow SSA measurement, which is more fully described in [Gallet et al. \(2009\)](#), hereafter GDZ09. Collimated radiation from diode lasers strikes a snow sample flush with the sphere bottom. After multiple scattering, the diffuse radiation striking an InGaAs photodiode is converted to a voltage. The photodiode sits behind an optical baffle to screen-out specular reflection. The voltage-to-reflectance conversion is calibrated by measurements of reflectance standards (Sphere Optics Inc.) made before and after snow sample measurement. Our experiments used two diode-lasers (635 and 1310 nm) to measure the visible and NIR snow albedos, A_{635} and A_{1310} , respectively.

2.1 Models

Snow reflectance is primarily determined by its specific surface area (SSA), the surface-area-to-mass ratio ([Grenfell and Warren, 1999](#)). SSA is inversely related to the optically effective snow grain size, aka effective radius r_e . Snow NIR reflectance is highly correlated with SSA ([Domine et al., 2006](#)), and is insensitive to small amounts of impurities. GDZ09 describe how we calibrate 1310 nm reflectance A_{1310} to SSA measured by methane adsorption techniques.

Snow albedo simulations were performed with the Shortwave Narrow Band (SWNB2) radiative transfer model [Zender \(1999\)](#); [Zender and Talamantes \(2006\)](#). SWNB2 solves for all radiant quantities at 10 cm⁻¹ spectral resolution from 0.2–5.0 μ m using the discrete ordinates technique. Ice refractive indices at 635 and 1310 nm values are as in [Warren and Brandt \(2008\)](#). Snow samples were treated as plane-parallel and homogeneous (constant density with depth). Edge effects are discussed below.

Model inputs include the snow density (measured), snow SSA (estimated from 1310 nm reflectance as in GDZ09), and the geometric standard deviation σ_g of the snow grain size distribution. [Grenfell and Warren \(1999\)](#) found $\sigma_g = 1.6$ is typical at the South Pole, and [Flanner and Zender \(2006\)](#) showed $\sigma_g = 2.3$ best fits observed grain-size evolution for young snow, and [Gallet et al. \(2009\)](#) found $\sigma_g = 1.4$ best matches reflectance measurements at $\lambda = 1550$ nm. The modeled reflectance for $\sigma_g = 2.3$ is greater than that for $\sigma_g = 1.4$ by about 0.8% and 1.1% for $\lambda = 635$ and 1310 nm, respectively. This study uses $\sigma_g = 2.3$ for consistency with the SNICAR model.

Our treatment of BC optical properties is identical to [Flanner et al. \(2007\)](#). Spectral variation of the BC refractive index is from [Chang and Charalampopoulos \(1990\)](#). A lognormal BC size distribution is assumed with number-median radius 50 nm and geometric standard deviation $\sigma_g = 1.5$. Tuning the BC density to 1322 kg m⁻³ leads to a mass absorption coefficient $\psi = 7500$ m² kg⁻¹ at $\lambda = 550$ nm, in agreement with the central estimate of [Bond and Bergstrom \(2005\)](#). We perform sensitivity studies to the effects of sulfate coatings on BC. These coatings increase the mass absorption coefficient relative to uncoated BC of the same mass by a factor of about 1.6 ([Bond et al., 2006](#)).

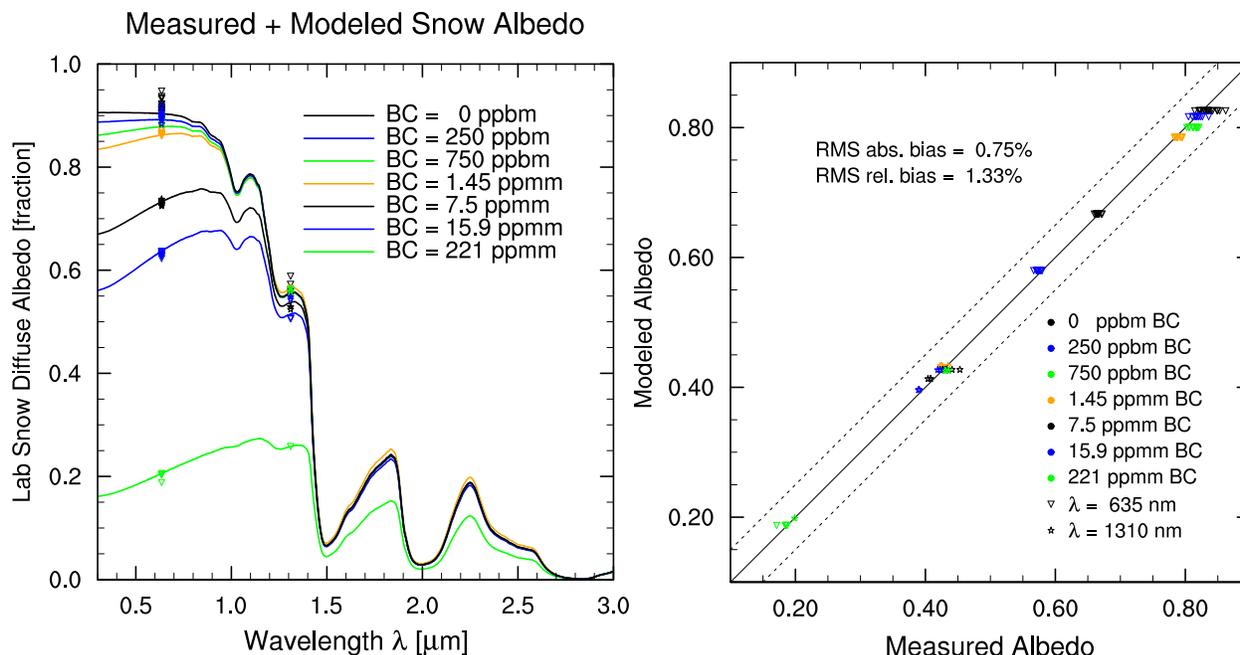


Figure 1: Measured and modeled snowpack reflectance for varying soot (BC) concentration.

3 Results

We report here reflectance measurements from $N = 97$ individual snow samples. Samples were either clean (not doped) or doped with one of six BC mixing ratios q_{BC} between 250 ppbm and 221 ppmm. [Gallet et al. \(2009\)](#) describe calibration of the NIR albedo with DUFISSS. These calibration parameters have not yet been confirmed for visible albedos. Pending the completion of this, we have employed a simpler, preliminary calibration that results in the modeled data shown in Figures 1a and b. The preliminary calibration simply assumes, based on least squares fits, that the conversion between plane-parallel semi-infinite albedos and DUFISSS measurements (which are neither plane-parallel nor semi-infinite) and is accomplished by multiplying the model estimates by 0.91 and 0.77 for visible and NIR albedos, respectively. This method yields an RMS bias of 0.75% (absolute) to 1.3% (relative) between modeled and measured albedo. A key uncertainty, not yet tackle, is verifying how much BC mass was “lost” during the doping process and thus did not fully contribute to snow darkening.

A series of further simulations (not shown) was conducted to assess the effect of different model assumptions on the model-measurement disparity. We found that using specific surface area (inferred from 1310 nm reflectance) improves visible snow albedo predictions (at 635 nm). The model-measurement disparity also improved when we assumed externally mixed uncoated BC rather than externally mixed sulfate-coated BC.

4 Conclusions

We measured the change in visible snow albedo due to doping the snow with six differing amounts of BC. Using a preliminary calibration, the albedo was within $\sim 1\%$ of predictions throughout the measured range of BC concentration, from 250 ppbm to 221 ppm. These results must be considered preliminary pending completion of the visible albedo calibration, and verification of the BC mass which affected snow albedo.

Acknowledgments

Supported by NSF ARC-0714088, NASA NNX07AR23G, and CNRS. We thank M. G. Flanner, T. Grenfell, J.-L. Jaffrezo, T. Kirchstetter, and S. Warren. Download this manuscript from http://dust.ess.uci.edu/ppr/ppr_ZGD09.pdf.

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