

Abstract

Snow and glacier melt water contribute water resources to a fifth of Earth's population. Snow melt processes are sensitive not only to temperature changes, but also changes in albedo caused by deposition of particles such as refractory black carbon (rBC) and continental dust. The concentrations, sources, and fate of rBC particles in seasonal snow and its surface layers are uncertain, and thus an understanding of rBC's effect on snow albedo, melt processes, and radiation balance is critical for water management in a changing climate. Measurements of rBC in a sequence of snow pits and surface snow samples in the Eastern Sierra Nevada of California during the snow accumulation and melt seasons of 2009 show that concentrations of rBC were enhanced seven fold in surface snow ($\sim 25 \text{ ng g}^{-1}$) compared to bulk values in the snow pack ($\sim 3 \text{ ng g}^{-1}$). Unlike major ions which are preferentially released during initial melt, rBC and continental dust are retained in the snow, enhancing concentrations late into spring, until a final flush well into the melt period. We estimate a combined rBC and continental dust surface radiative forcing of 20 to 40 W m^{-2} during April and May, with dust likely contributing a greater share of the forcing than rBC.

1 Introduction

Most water resources in the Western US originate as mountain snow in higher elevations, and climate warming will likely lead to enhanced winter snowmelt and earlier springtime release (Bales et al., 2006). Quantitative understanding of processes that influence snowmelt and spring runoff is critical to economic growth and ecological sustainability in these areas, since water conveyance systems for storage, flood control, power generation, recreation, and agricultural uses are designed and managed to optimize capture of spring snowmelt from the winter snow reservoir. While theoretical, field, and laboratory studies have found that aerosol contaminants in snow reduce its albedo (Warren and Wiscombe, 1980; Conway et al., 1996; Painter et al., 2007), research on

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the importance of radiative forcing from aerosols on snowmelt and water supply is limited and recent (Painter et al., 2010). Such aerosols include refractory black carbon (rBC, also called soot) and continental dust.

Refractory black carbon particles incorporated in snow enhance snow aging and melt, contributing to snow-albedo feedback and climate forcing (Flanner et al., 2007; Hansen and Nazarenko, 2004; Jacobson, 2004). The graphitic portion (comprised of greater than 60% carbon) and primary absorbing component of carbonaceous aerosols (Goldberg, 1985; Guggenberger et al., 2008), rBC is the product of incomplete combustion of fossil fuels and biomass. These particles strongly absorb solar radiation in the visible wavelengths; when mixed with highly transparent snow crystals, even minuscule concentrations reduce snow albedo and enhance melt (Conway et al., 1996; Warren and Wiscombe, 1980). Unlike major ions which are preferentially released during initial melt (Berg, 1992; Meyer et al., 2006, 2009; Williams et al., 2001), retention of rBC and continental dust in snow is not well understood. The radiative forcing from rBC in snow is determined by its distribution in the snow pack, both after initial deposition and during snow metamorphism and melt (Flanner and Zender, 2006). These aerosols have atmospheric residence times ranging from days to weeks and can be transported across oceans. Therefore both local and remote sources of rBC may deposit sufficient rBC to Sierra Nevada snow to enhance snowmelt and surface temperatures (Hadley et al., 2007, 2010)

Here we use measurements of rBC, continental dust, soluble ions, and physical properties in a sequence of snow pits in the Eastern Sierra Nevada snow pack in 2009, together with radiation modeling, to: (1) characterize concentrations of rBC in Sierra Nevada snow, (2) investigate changes in concentration and movement of rBC during snow accumulation and melt, and (3) simulate surface radiative forcing from measured rBC and continental dust in snow during winter and spring using the Snow, Ice, and Aerosol Radiative (SNICAR) model (Flanner et al., 2007).

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2 Methods

2.1 Sample collection

Field studies were conducted at a long-term study site on Mammoth Mountain in the Sierra Nevada operated by the University of California, Santa Barbara (UCSB) and the US Army Cold Regions Research and Engineering Laboratory (Painter et al., 2000). The hydrology of the site is dominated by a deep, well-sintered dry snow pack deposited by wintertime frontal storms moving from the Pacific Ocean, subsequent spring melt and generally mild dry summers punctuated by sporadic local convective storms (Harrington and Bales, 1998). Snowmelt from the site ultimately drains to the Owens River and the Los Angeles Department of Water and Power water conveyance system. Eight snow pits were excavated and sampled during the 2009 water year, with four pits during the snow accumulation period at one month intervals, and four pits during the melt period at one week intervals (Fig. 1). Sampling extended from the surface to the base of the snow pack at 10-cm increments using a 1-l stainless steel density cutter. Temperature, density, and stratigraphy measurements were recorded at the depth corresponding to each sample (Sterle, 2010). Three to six surface snow samples ~ 2 cm deep also were collected during each pit excavation. The snow pack generally consisted of coarse-grain snow clusters with mean grain diameter of 2 mm, although measurements were not made consistently at each sampling. Weather events and snowfall counts were not continuously monitored. Snow pit sampling during the accumulation period was coordinated with UCSB-led field campaigns aimed at measuring evolving snow microstructure (Bair et al., 2009), which documented coarsening increases in inter-grain distances and bond sizes. Once isothermal snow pack conditions were reached (29 April), snow pit sampling was conducted at one-week intervals.

Continuously recorded acoustic depth gauge measurements showed that peak 2009 snow accumulation occurred in early May. Lysimeters at the snow-soil interface showed no melt water releases from the snow pack before 1 May but substantial releases after, indicating that samples collected earlier are from the accumulation period and

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those after 1 May are from the melt period. Once melt started, occasional samples of melt water draining the snow pack also were collected from a suite of eight 1 m² snow lysimeters to monitor release of rBC from the snow pack. After each sampling period, still frozen snow samples and unfrozen lysimeter samples were transported to the Desert Research Institute in Reno, NV and stored at -10 °C until analysis for rBC, rock-forming elements, and soluble ions.

2.2 Sample analysis

Concentrations of rBC were measured using an intra-cavity laser-induced single particle incandescence soot photometer (SP2; Droplet Measurement Technologies, Inc.) coupled to an ultrasonic nebulizer (A5000T; Cetac). The method is described in detail by Bisiaux et al. (2011). A similar analytical system has been used extensively for rBC determinations in ice cores for analyzing continuous flow (McConnell, 2010; McConnell et al., 2007) and discrete samples (Kaspari et al., 2011). For the discrete snow sample measurements used in this study, samples were melted at room temperature and immediately ultrasonicated for 15 min just prior to rBC analysis in the original Fisherbrand Whirlpak bags used to collect the samples (product number 01-002-53). Samples and calibration standards were injected into the ultrasonic nebulizer and thence the SP2 using a 2.0 ml flow injection loop made of 0.5 mm (ID) Teflon tubing. Calibration standards were made from commercially available rBC hydrosols (Aqua-Black 162, Tokai Carbon, Tokyo). In these SP2 measurements of rBC, particle mass determinations were restricted to a range of 0.45 to 70 fg. Examination of the size distribution suggests that the vast majority of the rBC particles fall within this size range. Based on replicate measurements, the estimated uncertainty in rBC measurements ranged from 5 to 13 %.

Aliquots also were collected for analysis of soluble ions and continental dust. Soluble ions (ammonium (NH₄⁺), nitrate (NO₃⁻), chloride (Cl⁻)) were measured in selected snow pit samples from the accumulation and melt periods with continuous flow analysis methods routinely used for ice core analyses (Röthlisberger et al., 1999). To investigate

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continental dust, aliquots from the upper 30 cm of the snow pit samples were acidified immediately after melting with 1 % ultrapure nitric acid. These samples were analyzed for a broad range of rock-forming and other elements using High Resolution Inductively Coupled Plasma Mass Spectrometry (Element2, Thermo-Fisher). To allow for acid leaching of the dust particles, we analyzed the samples two to three months after acidification.

3 Results

Table 1 summarizes the geometric means and ranges of concentrations of rBC, soluble ions, and continental dust during the accumulation and melt seasons.

3.1 rBC concentrations in surface snow and snow pack

Results were interpreted under the assumption that the measurements reasonably represent temporal changes in rBC in the snow pack during the 2009 accumulation and melt seasons. While not capturing spatial variability or even an entire snow season, this dataset offers a first look at the evolution of ambient (i.e., not manipulated) rBC concentrations through the snow accumulation and melt periods. Snow pack rBC concentrations showed little temporal variation during accumulation and melt in either the geometric mean concentration or the concentration range (Fig. 1). However, concentrations in surface snow (upper 2 cm of the snow pack) were somewhat higher than bulk concentrations during both snow pack accumulation and melt (Fig. 2a), and substantially higher during the melt season (with a decrease at the end of the melt season).

Average rBC concentrations remained approximately constant during the first three weeks of melt from 1 May to 23 May, and then sharply decreased by 75 % between 23 May and 30 May (Fig. 2b). Fluxes of rBC from the base of the snow pack were estimated from measurements of rBC concentration in melt water captured in snow lysimeters on 23 May and measured melt water flow rates in the lysimeters from 23 May

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to 30 May. Fluxes ranged from 126 to 404 $\text{ng m}^{-2} \text{d}^{-1}$ – similar to the average rBC loss rate of 110 $\text{ng m}^{-2} \text{d}^{-1}$ over the same period determined from changes in snow pack concentrations.

3.2 Soluble ion concentrations

5 Concentrations of soluble ions HNO_3^- , NH_4^+ , and Cl^- sharply decreased during the first week of melting and then continued to decline as the 2009 melt season advanced (Fig. 2b).

3.3 Continental dust in the top 30 cm snow depth

10 Continental dust was measured only in the top 30 cm of the snow pack using the same snow samples collected for rBC measurements. Samples were collected at 10 cm intervals for all sampling periods. Rather than measuring dry mass load which is most suitable for high dust concentration samples and is potentially influenced by organic substances in the snow (e.g., Painter et al., 2007), dust concentrations were determined directly from measured cerium concentrations using the abundance of cerium
15 in mean sediment of 83 $\mu\text{g g}^{-1}$ (Bowen, 1979). Such methods for dust concentrations based on elemental measurements are routinely used in ice core studies (McConnell et al., 2007b, 2008; Banta et al., 2008). Concentrations of continental dust and rBC measured in the upper 30 cm of the snow pack showed similar patterns (Fig. 2c), with little change in concentration during the first three weeks of the melt season but then
20 rapid flushing during the fourth week of May. While appreciable SWE remained, sampling concluded at the end of May because of (a) difficult logistics associated with reaching the site, (b) risk of samples melting in warm temperatures, and (c) general site disturbance from trampling and a full season of experimentation.

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4 Discussion

4.1 Retention and mobility of dust, rBC, and ionic chemical species

We expected that dust, rBC, and ionic species would be immobile in the snow pack prior to the melt season because there is negligible melt redistribution during accumulation at this elevation. Indeed, total mass of rBC and ionic species increased during the accumulation season as the snow accrued, and the patterns of rBC concentration with depth observed in the snow pits were consistent from month to month during the accumulation period (Fig. 1).

Measurements of soluble ions in the snow pits showed removal during the early stages of melt (Fig. 1b), consistent with previous studies showing that soluble ions are released from the snow pack preferentially during the melt season (Melack and Stoddard, 1991). This preferential elution differs for each ion, suggesting different distributions of ions in the aged snow pack, release processes, and flow pathways in snow. This early release of soluble ions from melting snow pack contrasts with the delayed release of insoluble rBC and continental dust. This retention of particulate material during the early stages of melt is consistent with previous studies (Conway et al., 1996; Meyer and Wania, 2008).

4.2 Surface snow pack trends

The observed higher concentrations in surface snow relative to bulk averages in the snow pits for all sampling periods indicate that dry deposition of rBC is important at the Mammoth Mountain study site during the 2009 snow season. However, increasing relative surface concentrations of rBC during the melt season suggest that: (a) as the snow pack depletes from the surface, much or all of the previously deposited rBC concentrates at the surface, (b) increased anthropogenic activity in the area or altered atmospheric boundary layer processes during spring increased dry deposition during the melt season, or (c) rBC concentrations within precipitation (wet deposition)

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were enriched during late spring. While atmospheric and other meteorological measurements are needed to resolve the relative roles of these processes, we find it unlikely that melt-season precipitation became sufficiently enriched in rBC or that dry deposition increased during late spring to explain the 5-fold increase in surface rBC observed during the melt season (Table 1). Rather, because hydrophobic pollutants associated with larger particles are known to accumulate near the snow surface and are released at the end of melting (Meyer and Wania, 2008), we hypothesize that part or all of the rBC originally in the snow pack remained on the surface as the snow pack melted (Clarke and Noone, 1985). The result was progressive increases in mean rBC and dust (Fig. 2b, c), as well as increases in both absolute (Fig. 2a) and relative surface snow rBC concentrations (Table 1) early in the melt season, followed by a rapid release later, thereby lowering both mean and surface concentrations from 17 May to 23 May (Fig. 2a–c).

4.3 Snow pack radiative forcing from rBC and dust

We evaluated the daily mean radiative forcing of the rBC and dust with the SNICAR model (Flanner et al., 2007, available in a web interface at <http://snow.engin.umich.edu/>), which applies 470 spectral bands between 0.3 and 5.0 μm . We considered different snow grain sizes and applied the measured vertical distributions of rBC in the entire profile and distributions of continental dust measured in the top 30 cm of snow water equivalent. The forcing (Fig. 3) represents the daily-mean of the instantaneous change that impurities (rBC + dust) cause in the solar energy absorbed by the snow pack, calculated with a 30 min timestep. We determined surface forcings for both clear-sky and cloudy conditions, and weighted them equally to produce an all-sky forcing. The surface-incident spectral flux distributions were computed with an atmospheric radiative transfer model, applying a low liquid cloud with 500 nm extinction optical depth of 20 for the cloudy condition. We applied hydrophilic rBC optical properties (Flanner et al., 2007) and measured optical properties and size distribution of dust from Asian outflow over the Pacific (Clarke et al., 2004). Dust forcing is highly sensitive to both the

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size distribution and absorptivity. Here, we applied a number median radius of 0.69 μm , surface area-weighted (effective) radius of 1.40 μm , and a visible (spectrally-constant) imaginary index of refraction of 0.006, leading to relatively weak absorption per unit mass of dust (the mass absorption cross-section of this dust is 11 $\text{m}^2 \text{kg}^{-1}$ at 500 nm).

During the melt period at the Mammoth Mountain study site, both rBC and continental dust forcings peaked on 17 May, in the range of 7 to 23 W m^{-2} and 16 to 35 W m^{-2} , respectively. These component-specific forcings represent the incremental change in absorbed energy caused by each component, relative to snow pack containing only the other impurity. The retained rBC and dust enhanced radiative forcing in the snow pack well into the melt period, with dust contributing 1.5-fold greater forcing than rBC. Although there are large uncertainties in dust properties, both rBC and dust are sufficiently abundant to affect spring melt in Eastern Sierra Nevada snow.

5 Conclusions

Temporal variability of rBC and continental dust concentrations measured in a series of snow pits in the Eastern Sierra Nevada of California in 2009 suggests that the total snow pack rBC and dust masses increased during the snow accumulation. Consistent with prior studies of particle behavior in melting snow packs (Meyer and Wania, 2008), rBC and dust were conserved in the snow pack well into melt season, in contrast to the major ionic species, thereby enhancing concentrations near the surface until a final flush of melt water near the end of May 2009 (Fig. 2a).

Snow pack radiation model simulations indicate that retained rBC and dust enhance radiative forcing in the Eastern Sierra Nevada's spring snow pack (Fig. 2b, c), with dust contributing a greater forcing than rBC (Fig. 3). This feedback between rBC concentration, radiative forcing, and melt has been observed on Himalayan Glaciers (Xu et al., 2009) and may be an important process affecting the albedo of snow packs and glaciers on a global scale. A caveat to this study is that dry deposition of rBC and dust to the snow pack during the melt season was not measured independently and

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Table 1. Measured concentrations of rBC, soluble ions, and continental dust.

	accumulation season (Jan to Apr)		melt season (May)	
	geometric mean	range	geometric mean	range
rBC mass, ng g^{-1}				
bulk snow pack	3	< 1–89	3	< 1–75
upper 2 cm	25	3–81	135	20–429
soluble ions, $\mu\text{eq l}^{-1}$ (10 cm depth integration)				
NO_3^-	8	2–43	6	< 1–17
NH_4^+	3	< 1–36	2	< 1–7
Cl^-	34	7–135	28	9–110
continental dust mass, $\mu\text{g g}^{-1}$ upper 30 cm	12	3–53	12	1–44

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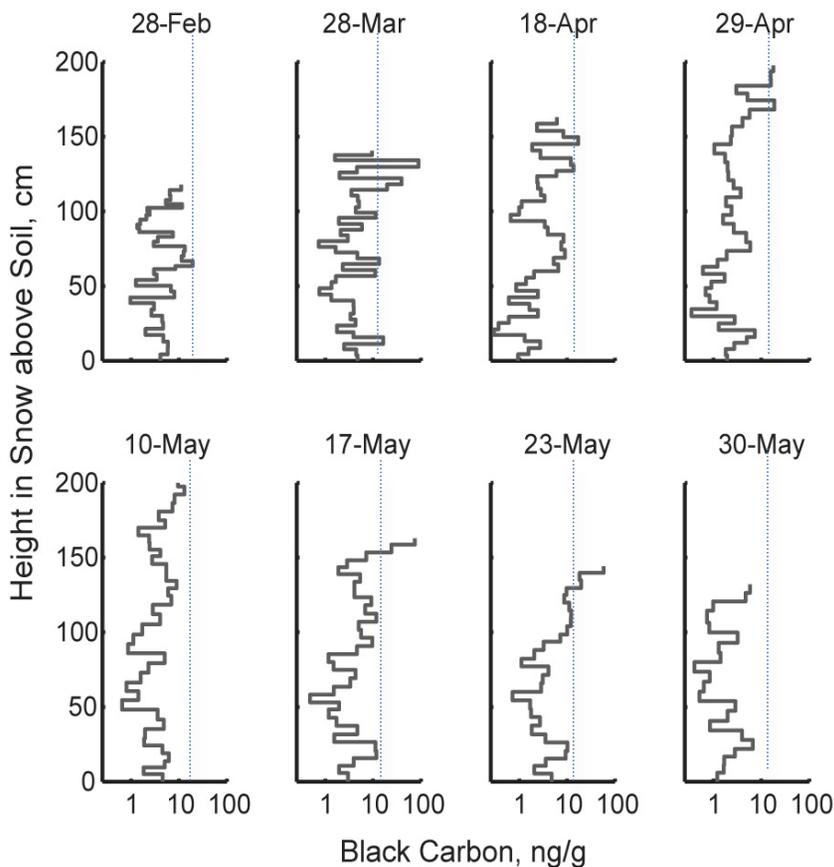


Fig. 1. Mammoth Mountain rBC concentrations (ngg⁻¹) versus height in snow above soil (cm water equivalence) in 2009 snow pit profiles.

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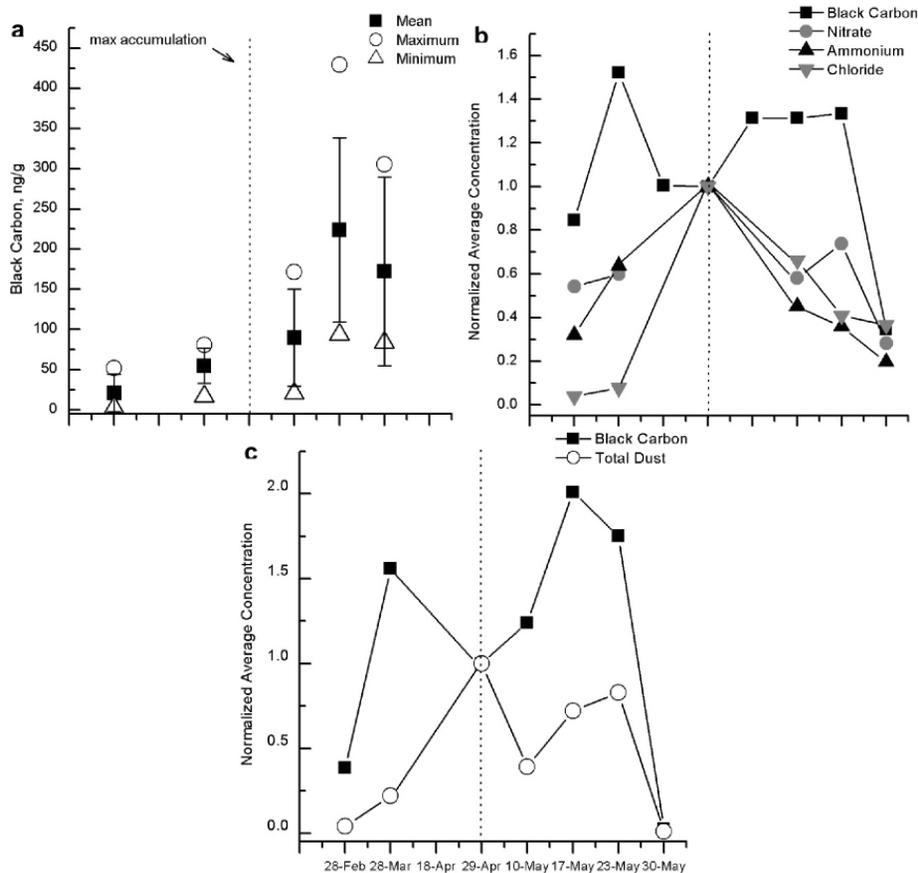


Fig. 2. (a) rBC concentration (ng g⁻¹) in surface (2 cm thick) snow samples, $n_{max} = 6$; (b) snow pit concentrations normalized to those at maximum accumulation when the snowpack became isothermal and melt water started to drain; (c) rBC and dust concentrations in the top 30 cm of snow pack normalized to those at maximum accumulation.

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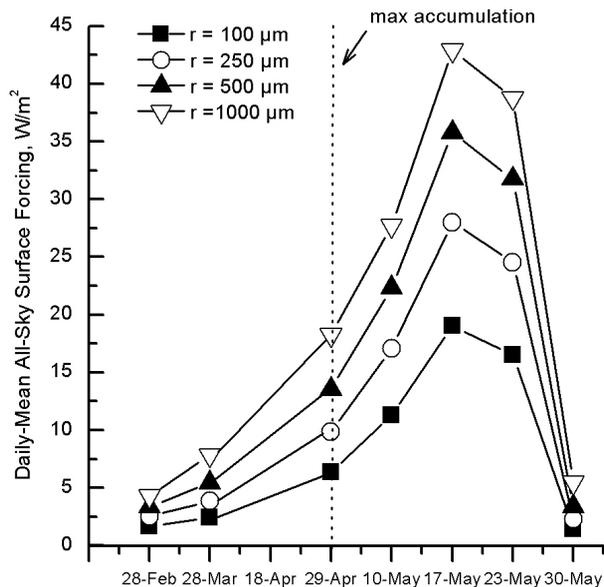


Fig. 3. Daily mean “rBC + dust” all sky (equally-weighted clear and cloudy conditions) surface radiative forcing with different snow effective radii based on the vertical distribution of rBC concentrations in entire snow pack profiles measured in 2009 at Mammoth Mountain, and continental dust measured in the top 30 cm of snowpack.

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