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**Black carbon
deposition on the
Sierra Nevada snow
pack**

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Measured black carbon deposition on the Sierra Nevada snow pack and implication for snow pack retreat

O. L. Hadley¹, C. E. Corrigan², T. W. Kirchstetter¹, S. S. Cliff³, and V. Ramanathan²

¹Lawrence Berkeley National Laboratory, Environmental Energies and Technologies Division, Berkeley, CA, USA

²Scipps Institute of Oceanography, University of California San Diego, La Jolla, CA, USA

³Department of Applied Science, University of California Davis, Davis, CA, USA

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Correspondence to: O. L. Hadley (olhadley@lbl.gov)

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Modeling studies show that the darkening of snow and ice by black carbon (BC) deposition is a major factor for the rapid disappearance of arctic sea ice, mountain glaciers and snow packs. This study provides one of the first direct measurements for the efficient removal of black carbon from the atmosphere by snow and its subsequent deposition on the snow packs of California. The early melting of the snow packs in the Sierras is one of the contributing factors to the severe water problems in California. BC concentrations in falling snow were measured at two mountain locations and in rain at a coastal site. All three stations reveal large BC concentrations in precipitation, ranging from 1.7 ng/g to 12.9 ng/g. The BC concentrations in the air after the snow fall were negligible suggesting an extremely efficient removal of BC by snow. The data suggest that below cloud scavenging, rather than ice nuclei, was the dominant source of BC in the snow. A five-year comparison of BC, dust, and total fine aerosol mass concentrations at multiple sites reveals that the measurements made at the sampling sites were representative of large scale deposition in the Sierra Nevada. The relative concentration of iron and calcium in the mountain aerosol indicates that one-quarter to one-third of the BC may have been transported from Asia.

1 Introduction

The effect that black carbon (BC) aerosols have on snow and ice reflectivity, and the subsequent impact on global ice extent and climate, has been a topic of research since the late seventies and early eighties (Chylek et al., 1983, 1984; Clarke and Noone, 1985; Warren and Wiscombe, 1980, 1985; Wiscombe and Warren, 1980). Studies showed that trace amounts of aerosol impurities in the snow can noticeably reduce the amount of sunlight reflected by snow and ice (Clarke and Noone, 1985; Warren and Wiscombe, 1980, 1985; Wiscombe and Warren, 1980). Reductions in snow and ice reflectivity and extent may substantially affect the global climate and exacerbate the

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warming effect due to greenhouse gases. For example, changes in global snow and ice cover account for over 50% of the observed fluctuations in the planetary albedo (Qu and Hall, 2005), an important variable in the global radiation budget (Ramanathan et al., 1989).

5 BC directly impacts snow albedo by absorbing solar radiation that would otherwise be reflected back to the atmosphere by the ice grains. Both the concentration of BC in snow and the size, or “age” of the snow crystals determines the degree of albedo reduction. Other factors include solar angle incident on the snow pack, surface cover (forest vs. meadow or rock), and snow depth. For BC concentration in snow of 10 ng/g and snow grains with an effective radius of 100 μm , models predict an albedo reduction of roughly 1% in the visible spectrum (Clarke and Noone, 1985; Flanner et al., 10 2007; Grenfell et al., 2002; Hansen and Nazarenko, 2004; Jacobson, 2007). As the snow pack ages the ice crystals undergo metamorphosis and grow. Bigger ice crystals amplify the light-absorbing effect of the BC inclusions by more than a factor of three (Flanner et al., 2007). This strong positive feedback leads to ever larger albedo reduction and further accelerates BC induced snow melt in the spring.

Several field campaigns were dedicated to measuring the concentration of BC in snow and ice in the arctic (Clarke and Noone, 1985), Antarctica (Chylek et al., 1987; Warren and Clarke, 1990), and various locations in North America and Europe (Chylek 20 et al., 1987; Grenfell et al., 1981; Sergent et al., 1987). Measurements of BC in glacial ice cores in the Himalayas have provided a historical record of BC deposition to the high altitude glaciers during the last fifty years (Ming et al., 2008). These results showed an increasing trend in BC deposition in the Himalayas and suggested that BC may play a role in the observed shrinking of the Himalayan glaciers and ice fields.

25 In addition to the impacts that BC may have on the radiation budget and thus, climate, the results of the Himalayan study point to a more immediate concern. In many regions of the planet, large population centers and agricultural economies rely on glacial and snow pack run off for fresh water during the dry season. One of these regions is California. Snow packs in the Sierra Nevada and the Southern Cascade mountain

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ranges are a crucial source of fresh water for California's agricultural production, as well as for densely populated urban and sub-urban centers. Increases in the snow pack melt rate are of major concern, as they contain the primary reservoir of water used to water Central Valley crops during the dry summer months (Painter et al., 2007).

5 The current study was conducted to examine the concentration of BC aerosols in snow in California and the potential of these aerosols to reduce albedo and increase melt. Samples of falling snow and rain were collected at three locations in California (Fig. 1): Central Sierra Snow Laboratory (CSSL) in the Sierra Nevada, Lassen Volcano National Park (LAVO) in the Southern Cascades, and at Trinidad Head (THD) on the Northern California coast. LAVO and THD are remote sites with few regional or local, large BC emission sources. CSSL, located downwind of Sacramento and San Francisco, is a much more polluted site. Air samples were collected and analyzed to quantify the mass concentration of BC and several elements. The amount of BC measured in Sierra Nevada snowfall during this field study was placed in context with the results from several modeling studies previously conducted to determine potential impact of BC on snow albedo and melt rate. Analysis of the elemental composition of the particulate matter in precipitation and air provided information about the sources of the BC. 5-year time series of BC and aerosol elemental composition at three separate locations in mountainous Central and Northern CA were used to estimate the spatial and temporal variability in BC, dust, and total PM_{2.5} (mass of particles of diameter less than 2.5 microns) concentrations. Size resolved atmospheric particulate measurements were made at both CSSL and LAVO, and atmospheric concentrations of BC were tracked at LAVO.

25 Adding to local and regional BC emissions in California, transport of pollution from Asia to North America over the Pacific Ocean is well documented as being greatest in the late winter and early spring (Bertschi and Jaffe, 2005; Bey et al., 2001; de Gouw et al., 2004; Goldstein et al., 2004; Hadley et al., 2007; Heald et al., 2006; Park et al., 2005; Parrish et al., 2004). The contribution of transported pollution to total observed pollution on the west coast of the United States is greater in the mountains

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than in low lying valleys and coastal regions (Hadley et al., 2007; VanCuren et al., 2005). The spring transport of Asian pollutants coincides with the onset of spring snow melt in west coast mountain ranges, which raises the possibility that black carbon (BC), the main light-absorbing component of soot, transported from Asia and deposited in the mountains, may also contribute to BC contamination of the snow pack. The ambient aerosol measurements, coincident with the precipitation collection, were used to estimate the relative influence of regional vs. long-range transport of BC.

2 Methods

Precipitation samples were collected between late February and mid-April 2006 using EcoTech, automated rain water samplers (RWS) modified for both rain and snow collection. An external gauge signaled the RWS lid to open and begin collecting when a precipitation event began and to close when it was over. The RWS collected one sample for each day of precipitation. In the event of extreme precipitation events, excess water was diverted to an external drainage tube. To collect snow, the external tipping bucket and collection funnels were heated, melting the snow as it fell on the surface. The temperature inside the collectors was kept at 10 °C or ambient temperature, whichever was higher. After collection, the precipitation samples were kept frozen until they could be filtered and analyzed. The largest volumes of collected snow and rain water were filtered through Pallflex-Tissuequartz fiber filters and analyzed for BC content using a modified version of thermo-optical analysis (TOA) (Hadley et al., 2008). Using laboratory standards of pure water and BC, filtration efficiency of a single filter was found to be only 30%; however use of three filters in series, increased the BC capture efficiency to 95% ($\pm 4\%$) and was therefore used in this analysis. Lower volume samples were analyzed for Fe (iron) and Ca (calcium) concentration using x-ray fluorescence spectroscopy (XRF) (VanCuren et al., 2005).

In addition to the collection of precipitation, several aerosol measurements were made at LAVO and CSSL. A Magee Scientific 7-wavelength aethalometer, measuring BC concentration, and a 3-wavelength EcoTech M9003 nephelometer, measuring

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the light scattering coefficient, were operated at LAVO for the duration of the experiment. The nephelometer data were used primarily to correct the aethalometer data for scattering artifacts (Arnott et al., 2005). The corrected, daily averaged, aethalometer BC concentration compared favorably with the IMPROVE (Interagency Monitoring for Protected Visual Environments) EC data (Chow et al., 2001) at LAVO. The linear regression slope was 0.9 with a correlation of $R^2=0.64$. The average BC concentrations used in this regression were all below 300 ng/m^3 where the uncertainties associated with both measurement methods are relatively large ($\pm 100\text{ ng}$). Ambient aerosol measurements also included size resolved mass concentrations for over 26 elements at LAVO and CSSL (VanCuren et al., 2005). Elemental mass concentration was measured continuously in 3-h time increments for both fine and coarse mode particles. Fe and Ca were used in this study as markers for Western United States vs. Asian dust to estimate how much of the BC was of Asian origin (VanCuren et al., 2005).

3 BC concentration in California precipitation

The average BC concentration in rain at THD, and in snow water equivalent (SWE) at LAVO and CSSL, respectively, were 5.7, 5.3, and 6.9 ng/g (see Table 1 for daily measurements by location). Similar measurements made by Chylek et al. (1999) near Halifax, Nova Scotia showed that BC concentration in snow in rural Nova Scotia averaged $1.7\pm 0.83\text{ ng/g}$, while snow falling near Halifax averaged $11\pm 7.7\text{ ng/g}$ of BC, illustrating that even in remote locales, the BC concentrations found in the Sierra Nevada snow were often similar to snow falling in an urban environment. These results also support previous model predictions for BC concentration in snow and rain in California. The GATOR-GCMOM (Gas, Aerosol, Transport, Radiation, General Circulation, Mesoscale, and Ocean Model) predicted concentrations of BC at 3 to 6 ng/g of precipitation over California in February. Other model studies predict BC concentrations in snow in California mountains between 10 and 22 ng/g (Flanner et al., 2007; Qian et al., 2009).

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Significantly heavier precipitation events at CSSL removed nearly 2.5 times more BC at CSSL than at either LAVO or THD (Table 1). Thus, although CSSL was a more locally polluted site, the heavier precipitation diluted the BC concentration measured at CSSL. The mass of BC removed each day (M_d) in ng/cm^2 was calculated as

$$M_d = M_c \cdot P \quad (1)$$

where M_c is the measured mass concentration in ng/cm^3 (kg) of snow or rain water and P is the total measured precipitation (cm) for a given sample day(s). The average BC masses removed by individual precipitation events at THD, LAVO, and CSSL, respectively, were 10.0, 11.5, and $24.3 \text{ ng}/\text{cm}^2$. Due to the size of sample required for analysis ($>100 \text{ mL}$), only BC mass in rain and snow from heavy ($>10 \text{ mm}/\text{day}$) precipitation events when sufficient snow could be collected was measured. Thus these samples were likely more diluted than the average concentration of BC in falling snow and therefore may underestimate the average BC mass concentration in snow.

4 Impact of BC on solar heating and snow melt

Models of radiation transfer in and above snow were used (Clarke and Noone, 1985; Flanner et al., 2007; Grenfell et al., 2002; Hansen and Nazarenko, 2004; Jacobson, 2004, 2007), along with the NCAR CCM3_CRM (Community Climate Model 3 Column Radiation Model), to estimate the potential impact of the BC concentrations found in the snow pack of the Southern Cascades and Sierra Nevada mountains on surface radiative forcing. Models predict that the visible (from 400 to 700 nm) albedo of snow is reduced by roughly 1% for every 10 ng BC per gram of fresh snow, assuming a snow grain effective radius of $100 \pm 50 \mu\text{m}$ and internally mixed BC and snow (Fig. 2). In older snow packs containing larger ice grains, albedo reduction would be much greater. Using the NCAR_CCM3 CRM and the modeled albedo reduction corresponding to the BC concentrations found in the Sierra Nevada, an estimate of the subsequent surface radiative forcing at the snow pack surface was determined. The following inputs

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were used in the CCM3 CRM. Latitude was set at 38°N and the day of year was set at 90.3333 GMT corresponding to 31 March at 16:00 PST and a solar zenith angle of 64° to represent a diurnal average. Control albedo for pure snow was set at 0.98 in the visible with subsequent albedo reductions of 0.3% (−0.003) and 1.2% (−0.012), respectively. BC does not appreciably lower the albedo in the near-IR (Warren and Wiscombe, 1980), although larger snow grains absorb significantly more radiation between 800 and 3000 nm (Wiscombe and Warren, 1980), which presents potential for a strong positive feedback from BC induced melt. For the range of concentrations of BC measured in snow at LAVO and CSSL, 2.5 to 13 ng/g, the CCM3-CRM predicted surface forcing above the snow pack is between 0.7 and 2.8 Wm^{−2}. The forcing predicted here by the observed BC concentration in snow is comparable with the pre-industrial to present CO₂ radiative forcing of 1.6 Wm^{−2} (Forster et al., 2007), although only regionally applicable and subject to large variability over said region.

Qian et al. (2009) predicted comparable mixing ratios for BC in snow water equivalent (SWE) and a similar change in net shortwave radiation flux of 1 to 3 Wm^{−2} in the Sierra Nevada during March. Subsequent simulations of the effect on snow melt and water in the Sacramento – San Joaquin river basins showed significant decreases in SWE throughout the spring and early summer, as well as an increase in surface temperatures. Flanner et al. (2009) also found that similar mixing ratios of BC in SWE could account for 20 to 30% of the simulated negative perturbation in March–May snow cover in the Western United States. This study provides observational support for recent modeling studies suggesting significant impacts of BC to the decrease of snow packs in California.

5 Sources of BC in snow

The potential impact that the measured BC/SWE mixing ratio may have on California snowpack compels us to examine the probable sources of the BC found in the snow. We first investigate the relationship between the particulate in the atmosphere and in

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the snow to determine how BC is transported to the snowpack. Secondly, we look at relative contributions from local and regional sources of BC vs. long range transport from global emissions.

At LAVO, the average ambient concentration of BC during the six hours just prior to the onset of precipitation was highly correlated ($R^2=0.56$) to the total amount of BC removed during the subsequent event (Fig. 3a and b), which suggested that below-cloud scavenging of BC from the atmosphere was the primary source of BC in the snow. Ambient BC concentration during the heavy precipitation events (>10 mm) decreased to levels near or below the aethalometer's lower limit of detection (roughly 15 ng/m^3), indicating that nearly all of the BC in the air was removed by the snow prior to the accumulation of 10 mm. Thus the scavenging of BC in the atmospheric boundary layer by falling snow likely accounts for most of the BC found in the snow at LAVO. Similar analyses for CSSL and THD were not possible because ambient BC concentrations were not continuously measured at these sites.

Mass concentrations of Ca and Fe in the air and snow measured at both LAVO and CSSL were used to infer the source region of the BC observed in California's mountains. The mass ratio of Fe to Ca in $\text{PM}_{2.5}$ observed on the west coast of North America is an indicator of North American vs. Asian dust (VanCuren et al., 2005). Van Curen et al. (2005) found that dust samples in which the $\text{PM}_{2.5}$ Ca mass was equal to or exceeded the Fe mass characterized Asian dust well. In North American dust, the Fe mass fraction of the $\text{PM}_{2.5}$ tended to be twice that of Ca or higher. Results from several elemental Asian dust characterization studies showed that the mass ratio of Fe to Ca ranges from 0.5 to 1 in Asian dust and pollution aerosols measured in China, Korea, and Hawaii (Holmes and Zoller, 1996; Krueger et al., 2004; Park et al., 2007; Shen et al., 2007; Sun et al., 2005). A dust characterization study for California's San Joaquin Valley revealed that the Fe to Ca ratio is around 2 for agricultural dust, the dominant source of dust during the Northern California wet spring. Although south and east of LAVO there are several large dry lake beds and deserts that produce dust aerosol with a Ca content about 4 times greater than Fe (Chow et al., 2003; Labban et al., 2004);

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predominant surface winds and calculated back trajectories show that dust from the east rarely impacts the western slopes of the Sierra Nevada in the spring months.

Using the observed Fe/Ca ratio in the fine atmospheric particulate matter at LAVO and CSSL, an estimate of the relative contribution of Asian PM_{2.5} soil dust to the observed PM_{2.5} soil dust was established. When the Fe/Ca ratio was greater than or equal to 2, the fractional contribution was assumed to be zero, when the Fe/Ca ratio was less than or equal to 1, the fractional contribution was assumed to be 1, and finally for Fe/Ca ratios between 1 and 2, the fractional contribution was scaled linearly (Van-Curen et al., 2005). The total soil mass concentration was calculated using the XRF elemental data and Eq. 2 (Eldred et al., 1997; Malm et al., 1994).

$$\text{Soil} = 2.2 \text{ Al} + 2.49 \text{ Si} + 1.63 \text{ Ca} + 2.42 \text{ Fe} + 1.94 \text{ Ti} \quad (2)$$

With the exception of three instances, the Fe/Ca ratio found in the snow samples at both LAVO and CSSL compare well with the PM_{2.5} Fe/Ca ratios in the surface air (Fig. 4). Therefore we conclude that these surface aerosols were generally representative of the ambient aerosols in the boundary layer and subject to scavenging by the snow. Thus the sources of BC and other aerosols in the atmosphere can be applied to that in the snow.

The average Asian BC contribution to BC mass observed at LAVO was estimated using the Asian dust fraction. Previous analysis of aerosol composition exported from Asia in the spring showed that the average BC, or EC, mass was approximately equal to 7% ($\pm 1\%$) of the PM_{2.5} soil dust mass (Bates et al., 2004; Moon et al., 2008; Shen et al., 2007). In North Eastern China, the average ratio of BC mass to soil dust mass in spring 2005 was 7.5% (Shen et al., 2007). The average springtime PM_{2.5} BC/dust ratio measured at a background site in Korea between 2001 and 2003 was 8% (Moon et al., 2008). In March and April 2001, PM₁₀ aerosol measurements, taken between Hawaii and Japan on NOAA's R/V Ronald Brown, presented a BC to soil dust ratio of 6.2% (Bates et al., 2004). Assuming that a proportionate amount of Asian BC is transported along with the dust to North America, estimates of the relative contribution of Asian

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BC to the total observed BC at LAVO can be made for spring 2006. Given that dust particles tend to be more massive than BC particles and may settle out more quickly during intercontinental transit, the following estimate of the fractional contribution of Asian BC to total BC in the Sierra Nevada may be biased low.

This estimate is valid only for dust and BC concentration measured during March, April, and May, as the field studies upon which it is based also reported only seasonal averages. BC to dust ratio at greater temporal resolution is likely to be significantly more variable. The estimated average Asian BC contribution to total average observed BC during the spring of 2006 was 27 to 36%, about 13% lower than the CFORS model estimated contribution (40 to 50%) at this altitude and location in the spring of 2004 (Hadley et al., 2007); it is however, in close agreement with simulations of Asian BC transport to the Western United States made during the TRACE-P (Transport and Chemical Evolution over the Pacific) experiment (Verma et al., 2008), which showed that 25% of the BC over the Western United States in the spring was from Asian origin. The average contribution of Asian dust to the total dust in the Sierra Nevada during this same time period was 87%.

Although these measurements were made at only two locations, one in the Southern Cascades and one in the Sierra Nevada range, an inter-comparison of five years of aerosol data taken at three different IMPROVE (Interagency Monitoring of Protected Visual Environments) sites (Fig. 1) showed that the aerosol measurements at these locations are spatially and temporally representative of mountains in Northern California (Fig. 5). At all three locations, there is a clear annual signal in the Fe/Ca ratio with a minimum value at or below 1 in the late winter and early spring, coinciding with the observed peak in trans-Pacific transport, and a late summertime high around 2, when local emissions dominate. Concentrations of EC, $PM_{2.5}$ soil and $PM_{2.5}$ are highly correlated and similar in magnitude at all three sites. These data provide justification that the measurements made at LAVO in spring of 2006 may be generally applicable to mountain conditions in Northern and Central California and moreover that 2006 was not an unusual year compared with the previous five.

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6 Implications for future trends

The data reveal that BC concentrations in the Sierra Nevada snowpack are sufficient to perturb both snow melt and surface temperatures. The concentration of BC measured in the snow is consistent with recent model predictions for BC concentration in California mountain snow. The associated reduction in snow albedo and reduced snow packs in early spring snowpack has been shown by regional climate models to be significant. A five year time series of BC, fine dust, and PM_{2.5} elemental composition from LAVO and two other sites in the IMPROVE data network indicate that the results presented in this study are spatially and temporally representative of conditions in the Southern Cascades and Sierra Nevada Mountains in Central and Northern California.

It is significant that the average contribution of Asian BC accounts for roughly one quarter to one third of the BC observed in the snow pack at high elevation sites in Northern and Central California. Between 1988 and 2001, the annual average atmospheric BC concentration in the San Francisco Bay Area has decreased from 2 $\mu\text{g}/\text{m}^3$ to less than 1 $\mu\text{g}/\text{m}^3$ (Kirchstetter et al., 2008), while at the same time, BC emissions from Asia have risen dramatically (Novakov et al., 2003). The fractional Asian contribution of BC to snowpack contamination in Western North America can be expected to increase should this trend continue.

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Table 1. Black carbon mass concentration measured in falling snow, precipitation amount, and total BC mass removed by the precipitation event at Trinidad Head (THD), Lassen Volcano National Park (LAVO), and Central Sierra Snow Lab (CSSL).

Site and date	BC conc. (ng/cm ³)	Precip (cm)	Total BC (ng/cm ²) rem.
THD 26 Feb 2006	2.3±2.3	2.2	5.1
THD 27 Feb 2006	4.5±3.7	4.4	19.8
THD 2 Mar 2006	5.0±1.8	2.1	10.5
THD 5 Mar 2006	1.2±1.2	2.3	2.8
THD 6 Mar 2006	9.8±5.9	1.3	12.7
THD 8 Mar 2006	12.9±3.6	1.6	20.6
THD 9 Mar 2006	9.9±3.5	2.9	28.7
THD 13 Mar 2006	3.6±2.3	1.4	5.0
THD 16 Mar 2006	1.7±1.2	1.5	2.6
THD 23 Mar 2006	8.3±3.1	0.9	7.5
THD 25 Mar 2006	3.0±2.0	2.7	8.1
THD 30 Mar 2006	4.1±3.7	1.2	4.9
THD 31 Mar 2006	5.5±2.5	1.2	6.6
THD 2 Apr 2006	7.8±3.1	0.7	5.5
AVG	5.7	1.9	10.0
LAVO 2 Mar 2006	7.0±2.2	1.8	12.6
LAVO 6 Mar 2006	3.7±2.1	2.1	7.8
LAVO 7 Mar 2006	6.0±4.7	2.4	14.4
LAVO 16 Mar 2006	4.0±3.3	1.1	4.4
LAVO 23 Mar 2006	4.2±1.5	3.7	15.5
LAVO 31 Mar 2006	9.6±3.9	1.3	12.5
LAVO 3 Apr 2006	2.8±2.3	4.7	13.2
AVG	5.3	2.4	11.5
CSSL 14 Mar 2006	7.9±1.5	3.4	26.9
CSSL 17 Mar 2006	7.3±2.5	1.7	12.4
CSSL 24 Mar 2006	9.8±2.4	6.3	61.7
CSSL 25 Mar 2006	8.0±1.7	2.4	19.2
CSSL 28 Mar 2006	11.2±4.5	3.7	41.4
CSSL 31 Mar 2006	5.8±1.6	1.9	11.0
CSSL 2 Apr 2006	2.9±1.9	5.5	15.9
CSSL 3 Apr 2006	2.5±2.5	2.4	6.0
AVG	6.9	3.41	24.3

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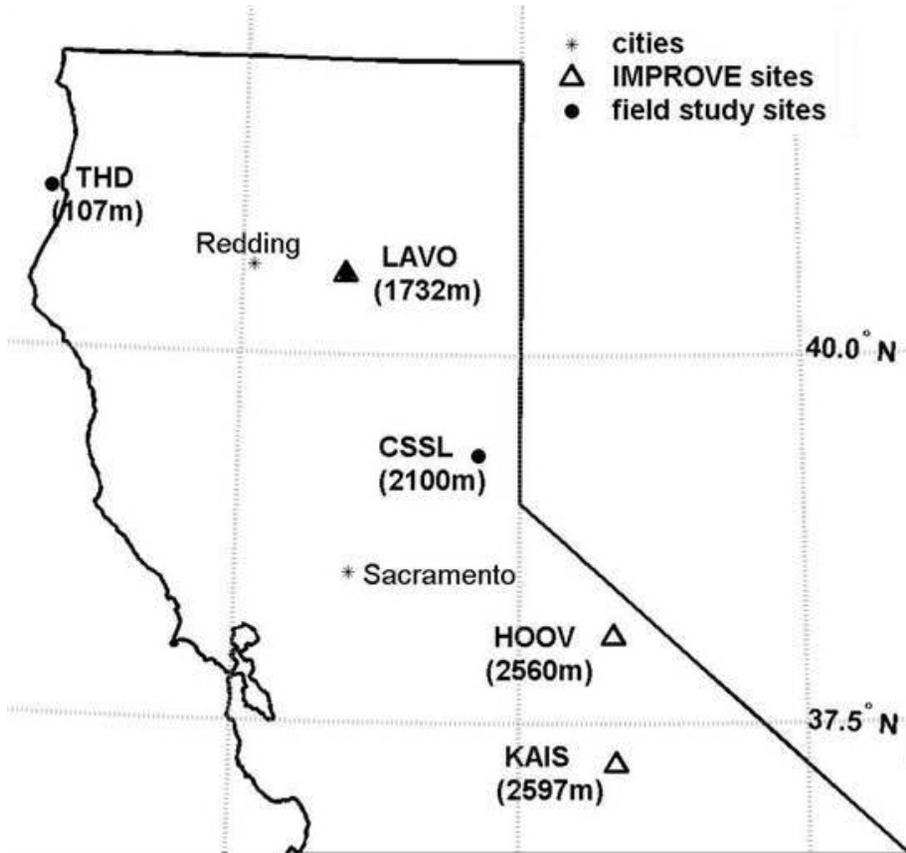


Fig. 1. Field study sites (circles) and IMPROVE sites (triangles) used in this study. LAVO is both an IMPROVE and field study site. The elevation is shown in parenthesis.

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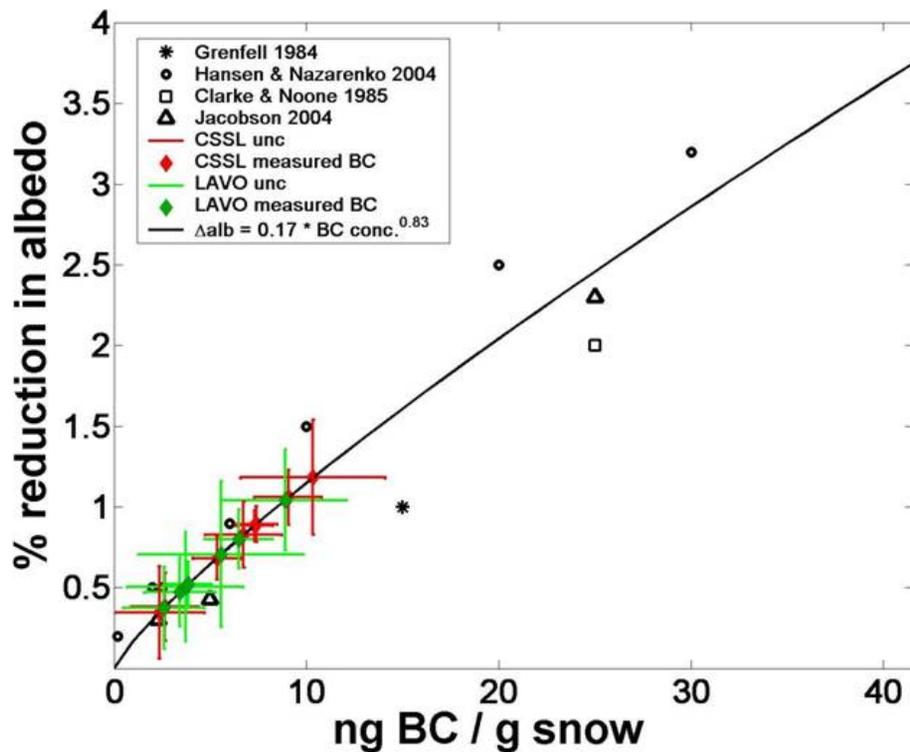


Fig. 2. Model results for BC induced albedo reduction on snow and subsequent potential for measured BC concentrations in falling snow at LAVO and CSSL to reduce snow surface albedo.

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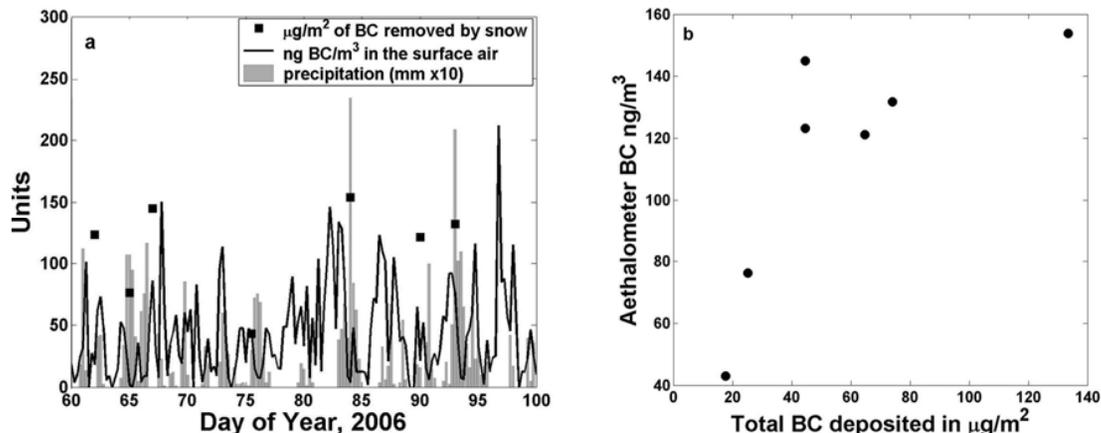


Fig. 3. (a) BC ng/m³ in the surface air (black line), mm of snow depth accumulated in 6-h increments (light grey bars), total BC μg/m² (units converted from ng/cm² to fit on figure) deposited by the falling snow during a 24 to 48-h period depending on timing of the snowfall (black squares). (b) Correlation of BC removed in a precipitation event to average ambient concentrations during the 6-h prior to the event. $R^2=0.56$. Data are from LAVO.

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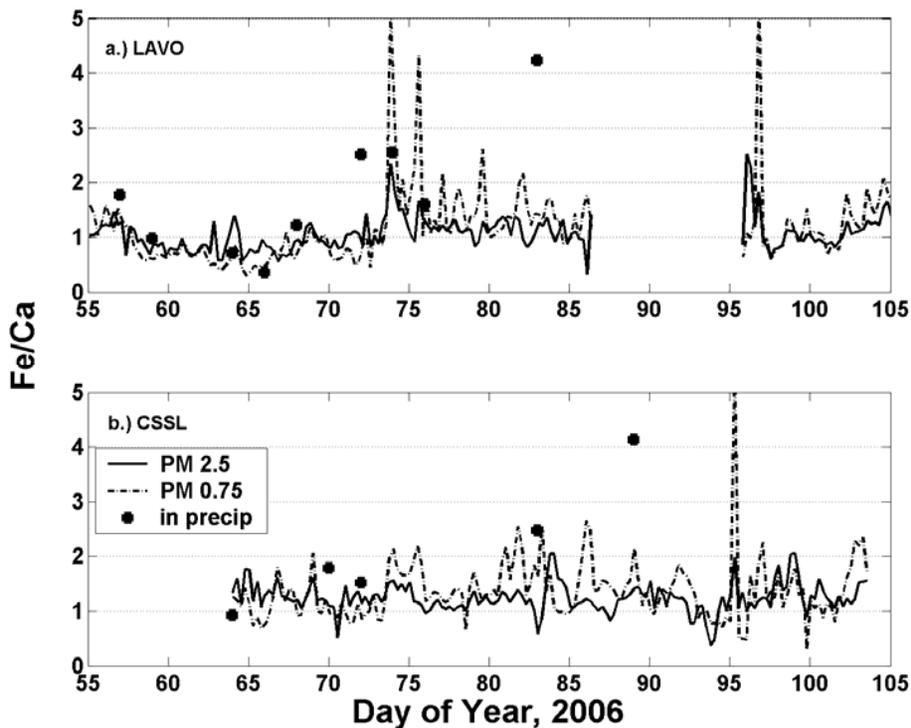


Fig. 4. Fe/Ca ratios for PM_{2.5}, PM_{0.75} ambient particulate matter, as well as in the precipitation, at (a) LAVO and (b) CSSL.

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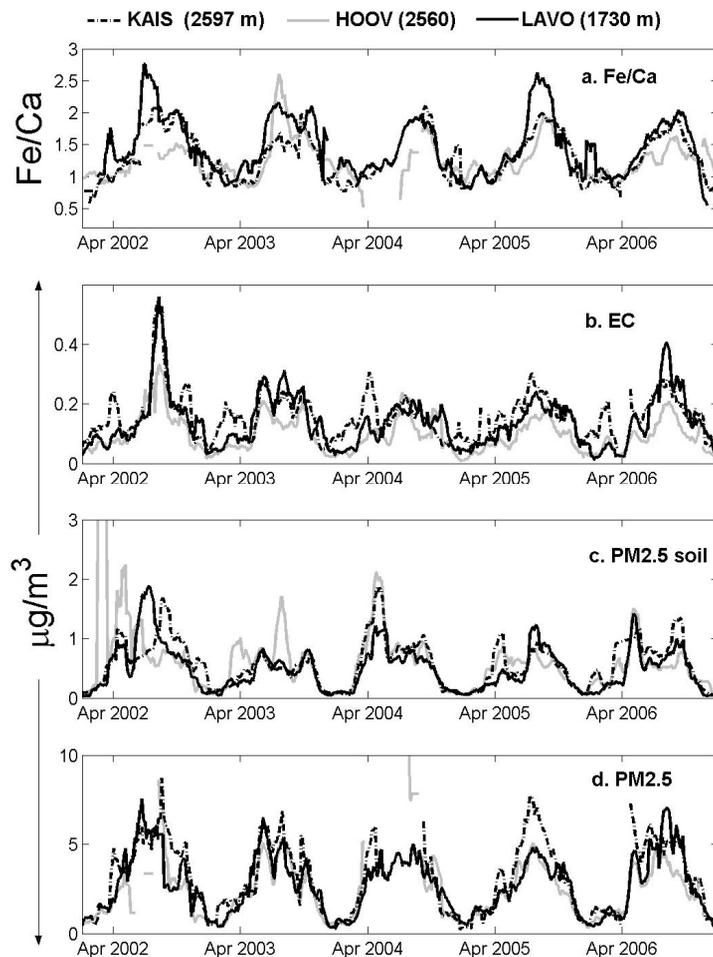


Fig. 5. Aerosol data from three IMPROVE (Interagency Monitoring for Protected Visual Environments) sites in Northern CA.

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