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Increasing aeolian dust deposition to snowpacks in the Rocky Mountains inferred from snowpack, wet deposition, and aerosol chemistry

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HIGHLIGHTS

- Snowpack calcium can be used as a surrogate for aeolian dust deposition to snow.
- Aeolian dust deposition to snow increased 81% in the southern Rockies during 1993—2014.
- Snowmelt timing accelerated 7—18 days over this time period in the Rockies, primarily due to changes in snowfall and dust deposition.

ARTICLE INFO

ABSTRACT

Mountain snowpacks are a vital natural resource for ~1.5 billion people in the northern Hemisphere, helping to meet human and ecological demand for water in excess of that provided by summer rain. Springtime warming and aeolian dust deposition accelerate snowmelt, increasing the risk of water shortages during late summer, when demand is greatest. While climate networks provide data that can be used to evaluate the effect of warming on snowpack resources, there are no established regional networks for monitoring aeolian dust deposition to snow. In this study, we test the hypothesis that chemistry of snow, wet deposition, and aerosols can be used as a surrogate for dust deposition to snow. We then analyze spatial patterns and temporal trends in inferred springtime dust deposition to snow across the Rocky Mountains, USA, for 1993—2014. Geochemical evidence, including strong correlations ($r^2 \geq 0.94$) between Ca^{2+}, alkalinity, and dust concentrations in snow deposited during dust events, indicate that carbonate minerals in dust impart a strong chemical signature that can be used to track dust deposition to snow. Spatial patterns in chemistry of snow, wet deposition, and aerosols indicate that dust deposition increases from north to south in the Rocky Mountains, and temporal trends indicate that winter/spring dust deposition increased by 81% in the southern Rockies during 1993—2014. Using a multivariate modeling approach, we determined that increases in dust deposition and decreases in springtime snowfall combined to accelerate snowmelt timing in the southern Rockies by approximately 7—18 days between 1993 and 2014. Previous studies have shown that aeolian dust emissions may have doubled globally during the 20th century, possibly due to drought and land-use change. Climate projections for increased aridity in the southwestern U.S., northern Africa, and other mid-latitude regions of the northern Hemisphere suggest that aeolian dust emissions may continue to increase, compounding the risk that climate warming poses to snowpack water resources in arid/semi-arid regions of the world.

1. Introduction

Mountain snowpacks are a critical resource in arid and semi-arid regions of the world, with more than one-sixth of the world's population relying on snowmelt for their water supply (Barnett et al., 2005). Snowpacks are a large natural reservoir,

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storing water during winter and releasing it during the summer and fall, when human and ecological demands are greatest (Barnett et al., 2008). Current and projected climate warming poses a substantial risk to snowpack resources, causing shifts in precipitation regime from snow to rain (Knowles et al., 2006), and shifting the timing of snowmelt towards earlier in the year (Cayan et al., 2001; Rauscher et al., 2008; Stewart, 2009; Clow, 2010). A recent study identified portions of southern Europe, the Middle East, and western North America as areas where climate-induced changes in snowpack resources may pose a substantial risk to summer/fall water supplies (Mankin et al., 2015).

Many of these areas are downwind from major sources of aeolian dust, which, when deposited on snow, can further accelerate snowmelt through increased absorption of solar energy (Conway et al., 1996; Painter et al., 2012; Skiles et al., 2012). Although uncertainties are high due to limited data, paleorecords suggest that desert dust emissions may have doubled across much of the globe during the 20th century (Mahowald et al., 2010), compounding the risk posed by climate warming to water resources in mountain areas globally.

Aeolian dust typically is fine-grained (silt-and clay-sized) material eroded from soil by wind; it can be transported at local, regional, and global scales, with concentration and grain size decreasing with distance (Bullard and Livingstone, 2009). Its mineralogy is usually dominated by quartz and feldspar, with lesser amounts of soluble salts, carbonates, and iron oxides (Loye-Pilot et al., 1986; Reheis et al., 2002). Dust deposition is highly episodic, typically occurring during major wind events in areas downwind from deserts and arid plateaus, such as the Alps, Himalayas, and Rocky Mountains (Laurent et al., 2006; Tanaka and Chiba, 2006; Kavouras et al., 2007). Globally, the largest sources of aeolian dust are in North Africa, the Middle East, and Central Asia (Mahowald et al., 2010). In North America, the largest dust sources are the Great Basin, Colorado Plateau, and Mohave and Sonoran Deserts in the southwestern U.S. and northern Mexico (Prospero et al., 2002; Tanaka and Chiba, 2006; Lawrence and Neff, 2009; Lawrence et al., 2010). Much of this area has sparse vegetation and is arid, receiving less than 120 mm of precipitation annually, and dust is carried predominantly eastward by prevailing winds (Prospero et al., 2002). Most dust is from natural sources, including playas and alluvial deposits (Reheis and Kihl, 1995; Tegen et al., 2004), but soil disturbance on desert, shrub, and grasslands can substantially increase soil erosion from those areas (Belnap and Gillette, 1997, 1998; Neff et al., 2005). Drought can cause increases in dust generation as well, due to its effect on soil moisture and vegetation coverage (Prospero and Lamb, 2003; Munson et al., 2011).

Studies in France, Italy, and Switzerland have documented high pH, Ca^{2+}, and alkalinity concentrations in "red" rain and snow from southerly storms tracking from the Sahara Desert, and the distinctive chemistry has been attributed to aeolian dust entrained by desert winds (Loye-Pilot et al., 1986; Schwikowski et al., 1995; Delmas et al., 1996; Rogora et al., 2004). Similar chemistry in rain and snow in the Rocky Mountains and Tien Shan (Asia) has been attributed to aeolian deposition as well (Williams et al., 1992; Clow and Ingersoll, 1994; Clow et al., 2002; Dong et al., 2009; Rhoades et al., 2010; Brahney et al., 2013). This chemical signature (high pH, Ca^{2+}, and alkalinity) may be explained by partial dissolution of carbonate minerals in dust during transport in the atmosphere or within melting snowpacks (Clow and Ingersoll, 1994; Delmas et al., 1996; Sala et al., 2008; Williams et al., 2009). Dissolution of carbonate dust can have the beneficial effect of neutralizing acid deposition, and in parts of southern Europe, it is estimated to reduce precipitation acidity by more than 50% (Draaijers et al., 1995; Psenner, 1999).

Meteorological and snow monitoring networks provide information required to track the effect of changes in climate on snowpacks. While dust deposition has been investigated through various research programs (e.g., Clow and Ingersoll, 1994; Painter et al., 2012; Landry et al., 2014; Skiles et al., 2015), there are no long-term (eg., >20 years), regional networks for monitoring aeolian dust deposition to snow using standardized methods (Miller et al., 2004; Bullard and Livingstone, 2009). Given the paucity of monitoring data on dust deposition to mountain snowpacks, and observations that dust may impart a distinct chemical signature on snow, we conducted a study to test the hypothesis that the chemistry of precipitation (snow and wet deposition) and aerosols could be used as a surrogate for dust deposition to snow. We then applied the methodology to evaluate spatial patterns and temporal trends in inferred dust deposition in the Rocky Mountains, USA (Rockies) during 1993–2014. The Rockies have experienced significant changes in snowmelt timing over the past several decades (Clow, 2010), as have other mountain ranges that are critical water supplies for arid/semi-arid regions of the northern hemisphere, such as the European Alps (Huss et al., 2009).

In this study, we used data from several long-term monitoring programs that have suitable geographic and temporal coverage for assessing dust deposition to snow in the Rockies. We have conducted annual chemical surveys of the snowpack at approximately 63 sites in the Rockies since 1993; these data cover the snowpack *accumulation* season, which typically is November—March. Dust deposition during the *snowmelt* season (typically April—May/June) was examined using data from the National Atmospheric Deposition Program/National Trends Network (NTN); the NTN has collected weekly composite samples of wet deposition in the U.S. since the late 1970s. Dust deposition to snow that occurs during non-precipitation events (dry deposition) was examined using data from the Interagency Monitoring of Protected Visual Environments (IMPROVE) program, which has monitored aerosol chemistry in the U.S. since the early 1990s. Data from a fourth long-term monitoring network, SNOTEL (SNOw TELemetry), were used to assess changes in *snowmelt timing*. The SNOTEL network is operated by the Natural Resource Conservation Service (NRCS), and provides data on snowpack depth and snow water equivalent (SWE) at approximately 730 sites across the U.S. Data from all four networks were combined in a multivariate statistical analysis to determine the relative influence of climate and dust deposition on snowmelt timing in the Rockies.

The paper is organized as follows: in Section 3.1, we evaluate the utility of using snowpack chemistry as a surrogate for dust deposition to snow. In Section 3.2, spatial patterns and temporal trends in snowpack chemistry across the Rocky Mountains are presented. In Section 3.3, we describe spatial and temporal patterns in wet deposition chemistry. In Section 3.4, trends in aerosol chemistry are presented. In Section 3.5, we analyze the influence of aeolian dust on snowmelt timing, and in Section 3.6, we discuss implications for snowpack water resources.

2. Methods

2.1. Study area

The study area is the Rocky Mountain region of the United States (U.S.), from northern Montana to northern New Mexico (http://co.water.usgs.gov/projects/RM_snowpack/; accessed 5/17/16). The Rocky Mountains form the headwaters for several major rivers in North America, including the Colorado, Rio Grande, and Missouri, which are critical water supplies for arid/semi-arid regions in the western U.S. This study area was selected because of its proximity to aeolian dust sources in the southwestern U.S., and because of

growing concern about the influence of climate change and aeolian dust deposition on snowpack water resources in the region. Climate models project increased drought frequency and severity in the southwestern U.S. over the next 50–100 years (Seager et al., 2007).

Ancillary snowpack and meteorological data were used from several research sites in Colorado, including Loch Vale and Niwot Ridge in the Front Range near Denver, and Senator Beck Basin in the southwestern part of the state.

2.2. Sampling and analytical methods

2.2.1. Snowpack

Depth-integrated samples of the seasonal snowpack have been collected by the U.S. Geological Survey (USGS) annually each spring since 1993 at approximately 63 sites in the Rocky Mountains to provide data on spatial patterns and temporal trends in snowpack chemistry for the Rocky Mountain region. None of the sites are on glaciers, and each sample represents total snow accumulation from the previous winter. Sampling sites range in elevation from 1588 to 3615 m, with 80% of sites between 2031 and 3373 m (see Table S1 for site coordinates and elevations). Samples were collected just prior to the time of maximum accumulation, which usually occurs in March. Mean daily air temperatures normally are below freezing throughout the winter; thus, mid-winter snowmelt at the sites is rare. Snowpack sampling methods and quality assurance procedures are described in detail in Ingersoll et al. (2002), and a brief summary is provided here. At each site, a snow pit was dug from the snow surface to the ground, and snowpack temperatures were recorded every 10 cm to verify that the snowpack had not begun melting. Snowpack density was measured at 10-cm increments using a 1-L density cutter and scale, and total snow water equivalent (SWE) was calculated by multiplying average density by snowpack depth. Median snow depth and SWE among all sites and years were approximately 1.5 m and 0.5 m, respectively. A depth-integrated sample of snow was collected by removing a column of snow from the north-facing wall of the snow pit using pre-cleaned plastic shovels, taking care to exclude the top 5 cm and bottom 10 cm of snow to prevent contamination from soil or forest litter. Field personnel wore powderless vinyl gloves, and all sampling equipment was cleaned prior to use by soaking in 18 M-ohm deionized water (DI), followed by triple rinsing with DI. Samples were placed in pre-cleaned 8-L Teflon bags, transported to the USGS laboratory in Denver, Colorado, on dry ice, and kept frozen at -20 °C until processing. Snow samples were melted at room temperature, filtered through 0.45-μm polyethersulphone filters, and analyzed for major element chemistry using methods designed for low-ionic strength water, as described in Turk et al. (2001) and Clow et al. (2002). Analytes included Ca^{2+}, Mg^{2+}, Na^+, K^+, NH_4^+, Cl^-, SO_4^{2-}, NO_3^-, pH, conductance, and alkalinity. Concentrations in field blanks were at or below the analytical detection limits, which were ≤ 1 μeq l^{-1} for all ions. Data are available at http://co.water.usgs.gov/projects/RM_snowpack/index.html (accessed 2/15/2016) and through the USGS National Water Information System website (http://waterdata.usgs.gov/nwis/qw; accessed 6/16/16).

On February 14–15, 2006, a frontal storm with high winds caused a major dust event in Colorado, depositing a prominent layer of aeolian dust on snowpacks across the western half of the state. This storm produced peak winds of 107 mph (48.6 m s^{-1}) during the early morning hours of February 14 at the Loch Vale research site, and sustained high winds throughout the day. Other mountain sites in western Colorado, including Niwot Ridge and Senator Beck Basin, experienced similar high winds, although the timing of peak wind speeds varied geographically. The dust layer that was deposited was subsequently buried by new snow, but remained readily visible as a dark band of snow several cm thick for

the remainder of the snow season (Fig. 1). While conducting routine sampling during the annual snowpack chemistry survey, we collected separate samples of the dusty snow layer at 13 sites in Colorado to obtain data on the chemistry and dust content of the layer. Samples of the dusty snow layer were extracted from the pit wall using the equipment mentioned previously, and were analyzed for the same constituents as the depth-integrated samples, plus dust concentrations. Dust concentrations were determined gravimetrically using pre-weighed, baked glass-fiber filters. Our objective was to examine relations between Ca^{2+}, alkalinity, and dust concentrations to determine whether Ca^{2+} and/or alkalinity could be used as a surrogate for dust concentrations in snow. To obtain information on the origin of dust, back trajectories were analyzed for the storm using the HYSPLIT model (Draxler and Hess, 1997) for 1° grid cells in western Colorado. Trajectories were run backwards for 24 h starting at 0000 UTC (coordinated universal time) on February 15, with a starting height of 500 m above ground level.

2.2.2. Wet deposition

Although snowpack chemistry is useful for characterizing aeolian dust deposition to snow during the snow *accumulation* season, it is less useful after snowmelt has begun because carbonate minerals will partially dissolve and Ca^{2+} will be flushed from the snowpack as melt progresses. Thus, an alternative surrogate is needed for the snowmelt season. For this part of the study, we chose to use wet deposition chemistry, which is monitored at approximately 260 sites across the U.S. by the NTN. The objective of the NTN is to provide data for characterizing spatial patterns and temporal trends in precipitation chemistry to evaluate the effects of air pollution on the landscape. Weekly composite samples are collected using automated samplers with a plastic bucket that is open during precipitation events, and is covered between events
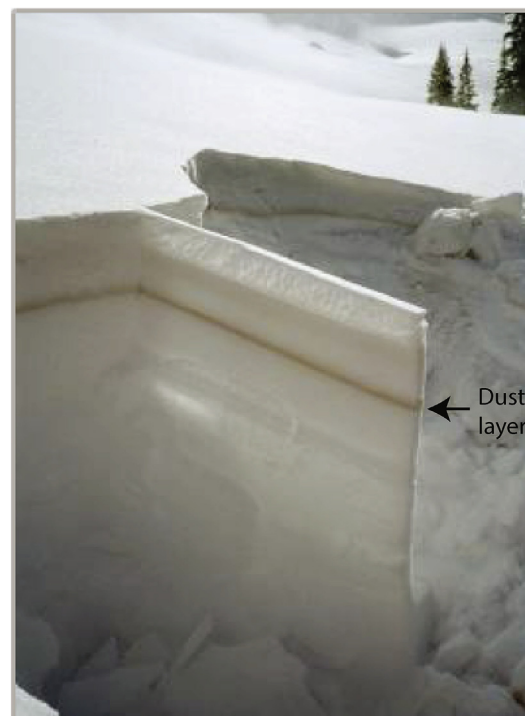

Fig. 1. Photo of snowpack in snow pit at Niwot Ridge (40.03°N, 105.54°W, 2900 m elevation) showing dust layer deposited on February 14–15, 2006. The dust layer is visible as a tan-colored band in the snow, approximately 15 cm below the snow surface.

(Peden, 1986). The NTN provides monthly, seasonal (spring, summer, fall, and winter), and annual summaries of precipitation-weighted mean (PWM) concentrations (e.g., μeq L^{-1}) and deposition (e.g., kg ha^{-1}) for all major constituents. Sample processing, analytical, and quality-assurance procedures are described at http://nadp.sws.uiuc.edu/lib/(accessed 5/19/16). In this study, we used spring season data from 31 NTN sites to analyze spatial patterns and temporal trends in Ca^{2+} concentrations in wet deposition in the Rocky Mountains for the same time period (1993–2014) and geographic area as the USGS snowpack network (see Table S2 for list of NTN sites used in analysis). The NTN defines spring season as March–May. We also analyzed trends in monthly concentrations to better define when trends (if any) were strongest (based on p-value and trend slope). Data were downloaded from the NTN web site (http://nadp.sws.uiuc.edu/ntn/; accessed 2/15/2016).

2.2.3. Aerosols

Aerosols and visibility are monitored at approximately 180 sites in the U.S. through the IMPROVE program. The primary objective of IMPROVE is to monitor visibility on protected federal lands, and identify chemical species responsible for visibility impairment (Malm et al., 1994). These species include sulfur and nitrogen compounds, organics, light-absorbing carbon, and aeolian dust (Malm et al., 1994). Aerosol samples of the <2.5-μm size fraction (PM2.5) and 2.5–10-μm size fraction (PM10) are collected over a 24-h period once every three days to obtain "daily values." A fine-soil component is calculated using the chemistry of the PM2.5 fraction, as described in Malm et al. (1994). We used aerosol data to analyze trends in the fine-soil component from 21 sites in the Rocky Mountains (1993–2014; Table S3); data were downloaded from the IMPROVE web site (http://vista.cira.colostate.edu/improve/data/IMPROVE/improve_data.htm; accessed 2/15/2016). For each site, monthly averages were calculated from the daily values for trend analyses. Operating procedures and quality-assurance reports for the IMPROVE network are available at http://vista.cira.colostate.edu/improve/Publications/publications.htm (accessed 5/19/16).

2.3. Statistical methods

2.3.1. Principal components analysis

Covariance among solutes in the snowpack data set was examined using principal components analysis (PCA). This technique is useful for simplifying complex data sets (e.g., many variables) to a smaller number of independent solute groups (components) that may be interpreted in terms of sources (Hooper and Peters, 1989). The strength of solute associations with a given component (source) is indicated by each solute's correlation with (or "loading" on) that component. In this study, the PCA was performed on a linear correlation matrix of the snowpack data, consisting of solute concentrations, site, and year. Components were rotated using the varimax method, and we retained components with eigenvalues greater than 1. All statistical analyses were performed using JMP version 12.1.0.

2.3.2. Trend tests

Trend tests were performed using the Mann-Kendall test, a non-parametric test that is resistant to outliers and makes no assumptions regarding the distribution of data (Helsel and Hirsch, 2002). Trend slopes were calculated as the median slope between all pairwise comparisons (Sen, 1968). Results of trend tests were evaluated for statistical significance at p ≤ 0.05.

For the snowpack data set, which had a relatively large number of sites, we used a modified form of the Mann-Kendall test called the Regional Kendall test (RKT). In the RKT, trends are tested on individual sites using Mann-Kendall and results are combined for

sites within a sub-region, thus gaining statistical power for detecting trends. This is important for short data sets with substantial inter-annual variability, such as the snowpack data. Sites were grouped into sub-regions based on close geographic proximity (usually coinciding with discrete mountain ranges), with 4–5 sites within each group. These criteria minimized subjectivity and provided similar statistical power for each group during trend analyses. The RKT was not used on the NTN, IMPROVE, or SNOTEL data sets because there were insufficient sites near the snowpack sites to warrant its use.

2.3.3. Stepwise multiple linear regression

To determine the relative influence of climate variables and dust deposition on snowmelt timing, stepwise multiple linear regression (MLR) was used to develop predictive models of snowmelt timing at SNOTEL sites nearest our snowpack monitoring sites (Table S4). A snowmelt timing index, SM50, was calculated for each SNOTEL site and year based on daily changes in SWE at the sites. SM50 refers to the day of the year when half of the snowpack has melted, using the date of maximum SWE as a reference (Clow, 2010).

Potential explanatory variables in the MLR analysis included: (1) monthly snowfall data from SNOTEL sites, (2) mean monthly air temperatures for 800-m grid cells corresponding to the SNOTEL sites, (3) snowpack Ca^{2+} concentrations, and (4) spring season Ca^{2+} concentrations in wet deposition. Data for the 62 SNOTEL sites were obtained at http://www.wcc.nrcs.usda.gov (accessed 2/15/2016). Air temperature data were obtained from http://cida.usgs.gov/thredds/catalog.html?dataset=cida.usgs.gov/topowx (accessed 2/15/2016). Snowpack Ca^{2+} concentrations were from our snowpack chemistry surveys, and wet deposition Ca^{2+} concentrations were from the NTN (see sections 2.2.1 and 2.2.2).

A brief summary of the MLR procedure is provided here; for additional details see Clow (2010). In the MLR, the variable that explains the most variance enters the model first. Variances of other variables are recalculated, and the one that explains the next greatest amount of variance enters the model next. This iterative procedure was used until the minimum Akaike information criteria (AIC) was obtained (Akaike, 1981). Multi-colinearity was evaluated using the variance inflation factor (VIF), with a threshold for exclusion of 4 (Hair et al., 2005).

3. Results and discussion

3.1. Can chemistry be used as a surrogate for dust in snow?

3.1.1. Solute associations in snow

Principal components analysis on the regional snowpack data set indicated that 84% of the variance in snowpack chemistry could be explained by four components, or sources (Table 1). The first and most important component, explaining 30% of the variance in the data set, had high positive loadings for solutes associated with aeolian carbonate dust, including Ca^{2+}, Mg^{2+}, and alkalinity (Table 1). This component also had a strong negative loading for H^+, consistent with neutralization of snowpack acidity by partial dissolution of aeolian carbonates in the snowpack (Psenner, 1999). The second component had high positive loadings for NO_3^- and SO_4^{2-}, the main anions associated with acid deposition (Turk et al., 2001). It is worth noting that H^+ was more strongly associated with the first component (carbonate dust) than with the second component (acid deposition). In previous PCAs on rain and snow, Hooper and Peters (1989) and Turk et al. (2001), found that H^+, NO_3^-, and SO_4^{2-} were all associated with an acid component. The negative association of H^+ with carbonate dust in the present study highlights the importance of aeolian carbonates in controlling

Table 1
Loadings and percentage of variance in snowpack chemistry explained by first four principal components. Bold indicates loadings ≥0.74 or ≤ −0.74.

	Carbonate dust	Acid deposition	Salt	Agriculture
Alkalinity	**0.91**	−0.05	0.25	0.01
Ca^{2+}	**0.80**	0.38	0.22	−0.06
Mg^{2+}	**0.74**	0.46	0.20	−0.15
K^+	0.50	0.31	0.00	−0.55
Na^+	0.18	0.07	**0.95**	0.03
NH_4^+	0.15	0.38	−0.01	**0.80**
Cl^-	0.22	0.10	**0.91**	−0.04
SO_4^{2-}	0.05	**0.86**	0.27	0.02
NO_3^-	0.04	**0.90**	−0.07	0.19
H^+	**−0.80**	0.36	−0.07	−0.20
Variance explained	30%	23%	20%	11%

snowpack acidity in the Rocky Mountains. The third component had high positive loadings for Na^+ and Cl^-, possibly from road salt or aeolian salt minerals blown from dry lake beds (Reheis and Kihl, 1995; Turk et al., 2001; Reynolds et al., 2003). The fourth component had a strong positive loading for NH_4^+, which we interpret as indicating emissions from feedlots or fertilized agricultural fields (Turk et al., 2001). To summarize, we interpret the four components as carbonate dust, acid deposition, salt, and agriculture.

3.1.2. Correlations of Ca^{2+}, alkalinity, and dust in snow deposited during a dust event

Back trajectory analysis of the February 14—15, 2006, dust event indicates that air parcels and dust originated primarily in northeastern Arizona and southeast Utah, and were carried up to 600 km northeast over western Colorado over the next 24 h (Fig. 2). These results are consistent with those of Skiles et al. (2015), who showed that dust events that impacted snowpacks in southwestern Colorado usually originated in the southern Colorado Plateau. These authors also found that dust events impacting the central Colorado mountains often had more westerly and northerly tracks (Skiles et al., 2015).

The snow layer that was deposited during the dust event of February 14—15, 2006, had a chemical signature strongly indicative of calcareous dust. Calcium and alkalinity concentrations were highly correlated ($r^2 = 0.998$, $p < 0.001$), and the slope of the regression was close to unity (Fig. 3a), indicating dissolution of calcite in snowmelt. Calcium and alkalinity concentrations also were strongly correlated with dust concentrations ($r^2 = 0.94$, $p < 0.001$), and the relation was non-linear (Fig. 3b). These data suggest that either Ca^{2+} or alkalinity could be used to estimate dust concentrations; however, alkalinity can be affected by strong acids from fossil fuel combustion (acid rain and snow), whereas Ca^{2+} is not. Thus, Ca^{2+} is likely to be a better surrogate for dust than alkalinity would be. The non-linear nature of the relation indicates a shift in mineralogy at higher dust concentrations, possibly due to changes in dust sources. Further research is needed to elucidate how the ratio of Ca^{2+} to dust concentrations varies with time and space. Additionally, it might be possible to develop more precise indicators of dust deposition by using a combination of major or trace elements (Lawrence et al., 2010; Dong et al., 2014); however, in the current study we restricted our analysis to constituents reported by the snowpack chemistry and NTN networks so that we could evaluate trends in inferred dust deposition.

3.1.3. Ca^{2+} concentrations in wet deposition during dust events

We tested the utility of using Ca^{2+} in wet deposition as a surrogate for dust by examining the chemistry of wet deposition that fell during several major springtime dust events, including those on February 14—15, 2006, and during March—April 2009. Table 2
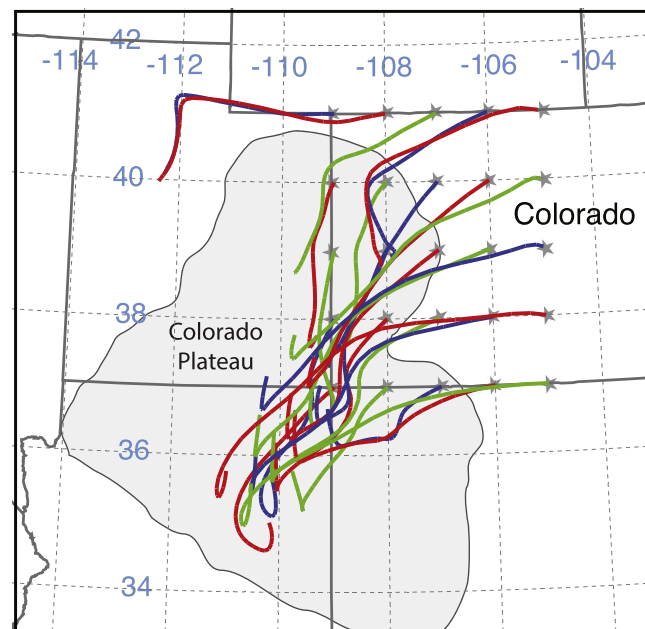

Fig. 2. Back trajectories calculated for 24-h period ending at 0000 coordinated universal time (UTC) on February 15, 2006, for 1° grid in western Colorado using HYSPLIT model (Hybrid Single-Particle Lagrangian Integrated Trajectory; http://ready.arl.noaa.gov/HYSPLIT.php; accessed 5/26/16). Stars indicate back-trajectory origins, and colors indicate individual trajectories. Solid grey lines are state boundaries. Dashed grey lines represent latitude and longitude.

shows Ca^{2+} concentration percentiles for weekly wet deposition samples collected after these events at 21 representative NTN sites in the Rocky Mountains. Results indicate that for the southern Rockies, post dust-event samples often had Ca^{2+} concentrations that ranked in the top 1% (99th percentile) of samples collected at that site during 2006 and 2009. Post dust-event samples also had high pH and Mg^{2+} concentrations, consistent with a carbonate dust signature. Samples collected in the northern Rockies after the dust events tended to have much lower Ca^{2+} concentrations, which fell within the normal range (Table 2), as expected given the greater distance of these sites from dust sources in the southwestern U.S. This might imply that either dust deposition is much lower in the northern Rockies, or Ca^{2+} is less useful as a proxy for dust in the north.

3.2. Spatial patterns and temporal trends in snowpack chemistry

Mean snowpack Ca^{2+} concentrations for 1993—2014 showed a strong latitudinal gradient, with relatively low concentrations at the northern sites (groups A—C) and central sites (groups D—G), and much higher concentrations at the southern sites (groups H—O), particularly those closest to the Colorado Plateau (Fig. 4a). These results are consistent with previous spatial analyses of Rocky Mountain snowpack chemistry covering the first 5 years of our study period (Turk et al., 2001; Clow et al., 2002). Snowpack Ca^{2+} concentrations in the southern Rockies (Fig. 4a) probably are influenced by aeolian carbonates because of their proximity to sources of calcareous dust in the southern Colorado Plateau (Clow and Ingersoll, 1994; Clow et al., 2002). In the southwestern U.S., playas have been identified as important sources of soluble salts and calcareous dust (Reheis and Kihl, 1995; Reheis et al., 2002). Multiple lines of evidence, including back trajectory modeling, remote-sensing imagery, and geochemical tracers, indicate that eastward transport of aeolian dust from the southwestern U.S. to
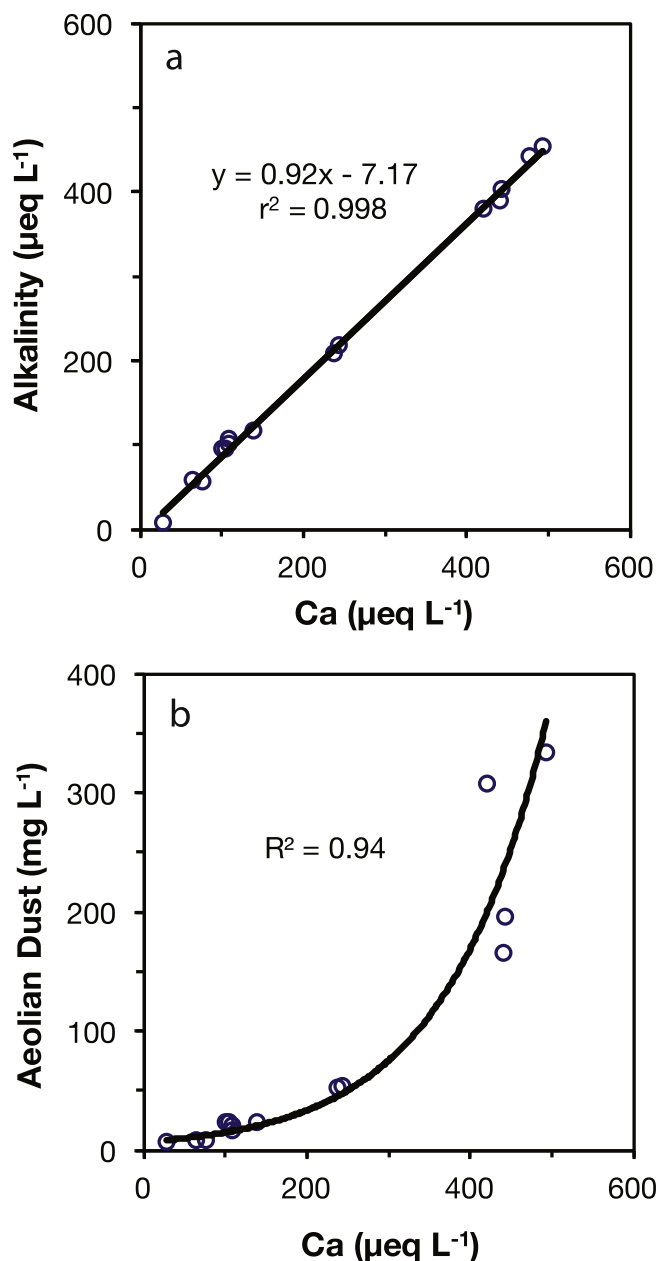
Fig. 3. Relations between (a) Ca^{2+} and alkalinity, and (b) Ca^{2+} and aeolian dust concentrations in dusty snow layer deposited on February 14–15, 2006. Data are from 13 snowpack sites in Colorado.

the southern Rocky Mountains is common (Reynolds et al., 2003, 2006; Painter et al., 2007; Lawrence et al., 2010; Skiles et al., 2015).

Our analysis identified strong upward trends in snowpack Ca^{2+} concentrations during 1993–2014 in the southern Rockies (0.23–0.83 µeq L^{-1} yr^{-1}), moderate upward trends in the central Rockies (0.14–0.23 µeq L^{-1} yr^{-1}), and weak or no significant trends in the northern Rockies (0–0.12 µeq L^{-1} yr^{-1}; Fig. 4b; Table 3). Average increases in snowpack Ca^{2+} in the southern, central, and northern Rockies were 81%, 65%, and 44%, respectively. Trends in snowpack alkalinity concentrations had a similar pattern, and were strongly correlated with trends in Ca^{2+} ($r^2 = 0.75$ for individual sites, $r^2 = 0.94$ for trends by group; $p < 0.001$ for both). The spatial pattern in trend magnitudes for snowpack Ca^{2+} and alkalinity are indicative of large increases in aeolian carbonate dust deposition in the central and southern Rockies. Possible explanations include

grazing (Neff et al., 2005; Belnap et al., 2009), agricultural tilling (Belnap et al., 2009), drought (Mahowald et al., 2010), increasing prevalence of high-wind events (Brahney et al., 2013), or increasing off-road vehicle use (Cordell et al., 2008).

It is noteworthy that trends in snowpack alkalinity were more strongly influenced by Ca^{2+} than by the anions associated with acid deposition (NO_3^- and SO_4^{2-}). Concentrations of NO_3^- declined at 8 of 15 groups, and SO_4^{2-} concentrations declined at 12 of 15 groups (Table 3); however, the magnitudes of the mean trends in NO_3^- (−0.07 µeq L^{-1} yr^{-1}) and SO_4^{2-} (−0.10 µeq L^{-1} yr^{-1}) were less than one-third of the mean trend in Ca^{2+} (0.32 µeq L^{-1} yr^{-1}).

3.3. Spatial and temporal patterns in wet deposition chemistry

As mentioned in Section 3.1.3, wet deposition chemistry provides an alternate (albeit indirect) method for examining spatial patterns and temporal trends in aeolian dust transport during the snowmelt season. Carbonate dust undergoes partial dissolution in the atmosphere during transport, generating Ca^{2+} and alkalinity, which reacts with H+ to neutralize acidity derived primarily from fossil fuel combustion (Loye-Pilot et al., 1986; Sequeira, 1993; Rogora et al., 2004; Jacobson and Holmden, 2006). Studies in southwestern Colorado and Japan have shown that ~70% of springtime dust events are associated with precipitation (Inomata et al., 2009; Lawrence et al., 2010), suggesting that wet deposition chemistry might provide a useful tool for tracking patterns and trends in dust deposition.

Average springtime Ca^{2+} concentrations in wet deposition for 1993–2014 increased from north to south (Fig. 5a), similar to the pattern in average snowpack Ca^{2+} concentrations. These patterns reflect the relative influence of aeolian carbonate dust on wet deposition chemistry in the Rockies, with sites in the south experiencing the greatest effect due to proximity to upwind dust sources in the Colorado Plateau (Turk et al., 2001; Clow et al., 2002).

Trends in springtime Ca^{2+} concentrations in wet deposition during 1993–2014 were strongest in the southern Rockies, with 60% of sites showing upward trends ranging from 0.38 to 1.34 µeq L^{-1} yr^{-1}, and no downward trends (Fig. 5b). In the central Rockies, 50% of sites had upward trends (0.57–0.74 µeq L^{-1} yr^{-1}), and there were no downward trends. Trends were weakest in the northern Rockies, where the only significant trend was downward at one site (−0.21 µeq L^{-1} yr^{-1}). Investigating the seasonality of these trends in more detail, an analysis of monthly PWM Ca^{2+} concentrations in wet deposition indicated that trend slopes were greatest during April; median Ca^{2+} increases at the 31 NTN sites during March, April, and May were 0.24, 0.41, and 0.27 µeq L^{-1} yr^{-1}, respectively.

Results from this study are consistent with those of Brahney et al. (2013), who documented large increases in annual Ca^{2+} deposition in the inter-mountain west during 1994–2010. The trends in annual deposition were largely driven by increases during springtime, and were correlated with the prevalence of high-wind events (>25.7 m s^{-1}; $r^2 = 0.53$). It should be noted that the very large increases in springtime Ca^{2+} deposition (1076%) reported by Brahney et al. (2013) reflect several large Ca^{2+} deposition years near the end of their record (2006, 2009, and 2010). More recent data from the Loch Vale NTN site show generally lower Ca^{2+} deposition values since 2010, although an overall upward trend is still evident (Fig. 6; $p < 0.04$). These observations highlight the episodic nature and high inter-annual variability in dust deposition, which is driven by a combination of high-wind events and varying drought conditions, vegetation coverage, and land use (Belnap and Gillette, 1998; Mahowald et al., 2003, 2010; Prospero and Lamb, 2003; Munson et al., 2011).

Table 2
Calcium concentration percentiles for weekly wet deposition samples collected after major springtime dust events of 2006 and 2009 at high-elevation NTN sites in the Rocky Mountains. [Percentiles indicate the sample concentration's rank within the statistical distribution for all weekly samples collected at a given site during 2006—2009. Blank cells indicate minimal precipitation during the week (no sample). Sites are listed in order by latitude from north to south. See http://nadp.sws.uiuc.edu/ntn/for map of sites].

NTN site ID	2/14/06—2/21/06	3/17/09—3/24/09[a]	3/24/09—3/31/09[a]	3/31/09—4/7/09[a]	4/21/09—4/28/09[a]
MT05		75	50	<50	75
MT07		75	<50	<50	50
WY08			<50	<50	50
WY98	<50	90	<50	<50	50
WY06	<50	97.5		<50	<50
WY02	<50	75	<50	<50	<50
WY00	90		50	75	90
WY95	75	95	50	75	95
UT08	<50		<50	<50	50
CO97	95	99	75	97.5	99
CO93	95	99	90	97.5	97.5
CO15	75	97.5	99	75	95
CO19	99		<50	99	97.5
CO98	99		75	99	
CO02	99	95	50	99	99
CO94	99		<50	95	95
CO92	75	97.5	75	97.5	99
CO08	90	97.5	90	99	99
CO10	50	99	75	99	99
CO96	97.5	99	97.5	97.5	99
CO91	75	97.5	90	97.5	

[a] Dust events recorded by observers with the Center for Snow and Avalanche Studies at the Senator Beck study site (http://www.codos.org/#dust-enhanced-runoff-classification; accessed 2/15/2016).
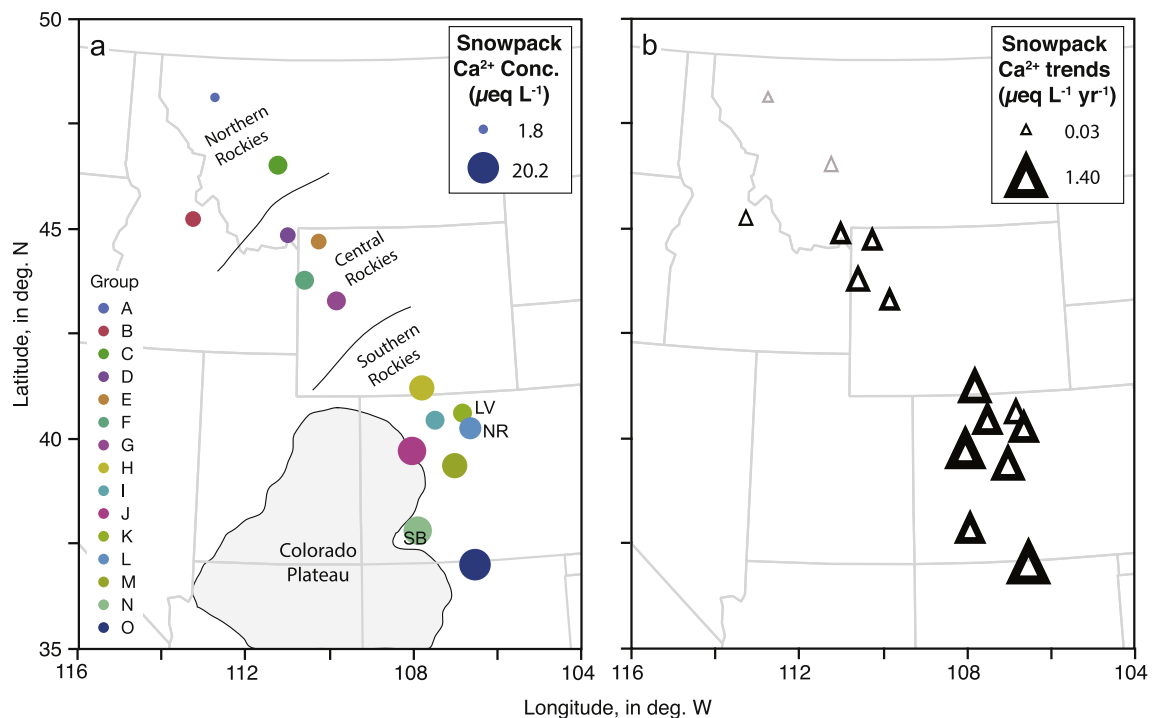

Fig. 4. Maps showing (a) spatial patterns in average Ca^{2+} concentrations in snowpack, 1993—2014, and (b) trends in snowpack Ca^{2+}, 1993—2014; groups with p ≤ 0.05 in black; those with p > 0.05 in grey. Abbreviations LV, NR, and SB refer to Loch Vale, Niwot Ridge, and Senator Beck research sites. Upward-pointing triangles indicate a positive trend slope; downward-pointing triangles indicate a negative trend slope. Symbol size indicates magnitude of trend. Curved black lines indicate boundaries between Northern, Central, and Southern Rockies regions. Northern Rockies includes groups A—C; Central Rockies includes groups D—G; Southern Rockies includes groups H—O.

3.4. Trends in aerosol chemistry

Although most dust events in the Rocky Mountains are associated with precipitation, approximately 30% are associated with dry deposition, suggesting that dry deposition may contribute substantially to total dust deposition flux (Lawrence et al., 2010). Trend analyses on the fine-soil component of aerosols from IMPROVE indicate few significant trends during most months, with the exception of March. During March, 8 of 21 sites showed upward trends, with most occurring in the southern Rockies (Fig. 7). These results are qualitatively consistent with our trend results on snowpack and wet deposition chemistry, which indicated upward trends in winter/spring Ca^{2+} concentrations in the southern Rockies. Timing differences in trends in wet deposition (greatest in

Table 3
Trends in snowpack solute concentrations, 1993−2014, by sub-region group[a]. Statistically significant trends (p ≤ 0.05) highlighted in bold. Units are µeq L^{-1} yr^{-1}.

Group	H$^+$	Alkalinity	Ca^{2+}	Mg^{2+}	Na$^+$	K$^+$	NH$_4^+$	NO$_3^-$	Cl$^-$	SO$_4^{2-}$
A	**−0.09**	**0.21**	0.02	0.00	0.00	0.00	0.02	**−0.05**	0.01	**−0.07**
B	−0.02	**0.26**	**0.12**	**0.03**	**0.05**	**0.04**	−0.01	−0.02	**0.04**	−0.01
C	−0.09	**0.39**	0.11	**0.04**	0.02	**0.03**	0.00	−0.05	0.01	**−0.13**
D	**−0.15**	**0.41**	**0.15**	0.01	**0.06**	0.01	0.08	**−0.07**	**0.04**	**−0.11**
E	**−0.14**	**0.33**	**0.14**	0.01	0.03	**0.01**	0.10	**−0.04**	0.01	**−0.05**
F	**−0.12**	**0.49**	**0.23**	0.04	**0.07**	0.02	0.21	−0.04	**0.02**	−0.05
G	**−0.11**	**0.43**	**0.15**	0.02	0.00	**0.02**	0.06	**−0.06**	0.01	**−0.11**
H	**−0.42**	**0.80**	**0.55**	**0.07**	**0.07**	**0.05**	0.11	**−0.13**	0.02	**−0.14**
I	**−0.50**	**0.76**	**0.36**	**0.05**	**0.04**	**0.03**	0.11	−0.05	**0.02**	**−0.19**
J	**−0.18**	**0.89**	**0.60**	**0.08**	**0.05**	0.02	0.05	**−0.09**	**0.03**	**−0.12**
K	**−0.28**	**0.53**	**0.23**	0.02	0.03	0.03	**0.07**	−0.07	0.01	**−0.13**
L	**−0.40**	**0.64**	**0.38**	**0.05**	0.02	**0.03**	0.09	**−0.11**	0.02	**−0.15**
M	**−0.19**	**0.76**	**0.53**	**0.09**	**0.05**	**0.04**	**0.06**	**−0.09**	**0.05**	**−0.05**
N	**−0.12**	**0.63**	**0.34**	**0.07**	0.01	**0.03**	**0.05**	−0.05	0.02	**−0.12**
O	**−0.25**	**1.40**	**0.83**	**0.17**	0.04	**0.15**	0.04	−0.06	**0.05**	−0.10
Average	−0.20	0.59	0.32	0.05	0.04	0.04	0.07	−0.07	0.02	−0.10

[a] See Fig. 4 for map of groups. Groups A−C, northern Rockies; D−G, central Rockies; H−O, southern Rockies.
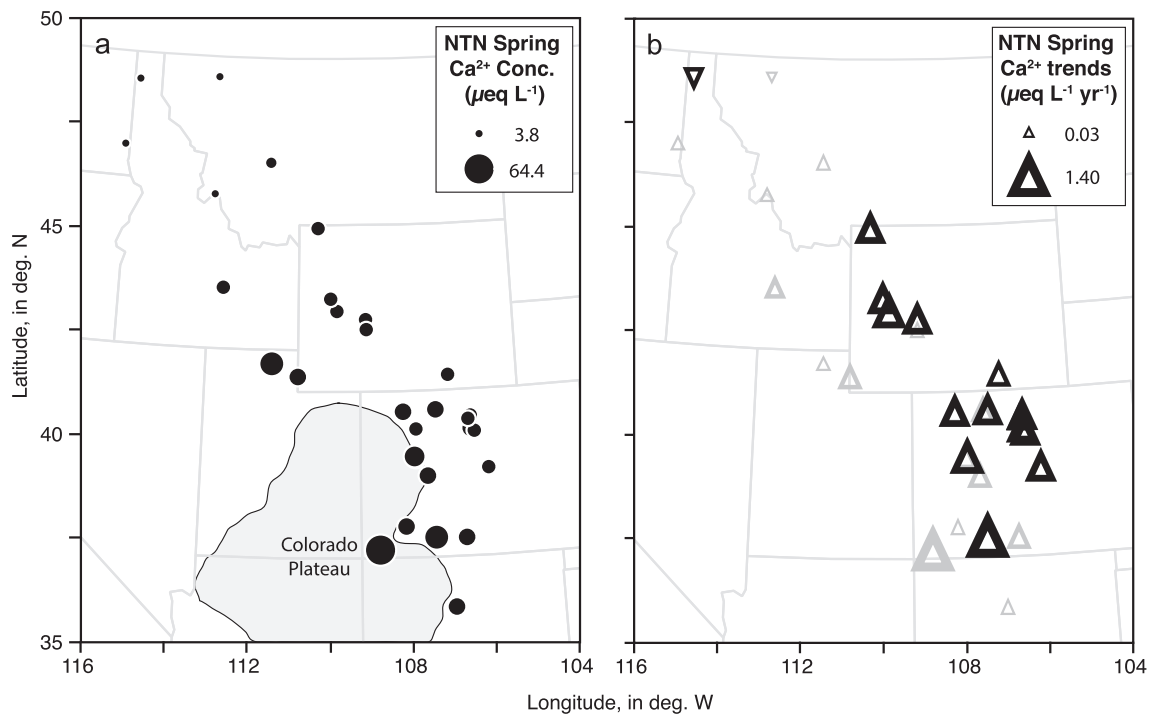
Fig. 5. Maps for springtime Ca^{2+} concentrations in wet deposition for 1993−2014 showing (a) spatial patterns in average concentrations, and (b) temporal trends. Symbols as in Fig. 4. Data are from National Trends Network (NTN).

April) and dry deposition data (March) may be indicative of differences in climatology of dust transport between these months. However, interpretation of the aerosol data is limited by the lack of continuous monitoring by IMPROVE, which samples on one of every three days (Malm et al., 1994). Because of the episodic nature of dust events, IMPROVE captures some events, but misses others. Additionally, IMPROVE only samples particles ≤10 µm in size, below the size of most regional aeolian dust (10−40 µm; Lawrence et al., 2010; Neff et al., 2013).

3.5. Influence of aeolian dust on snowmelt timing

During 1993−2014, snowmelt timing (SM50) advanced (became earlier) by 7−18 days in the southern Rocky Mountains, indicating shifts towards earlier snowmelt in this area (shown as downward arrows in Fig. 8a). Changes in SM50 in the central and northern Rockies were much less pronounced, and although most regression slopes were negative (indicating earlier melt), none of the trends were statistically significant. Trends in the timing of maximum SWE and the date of complete melting had similar patterns, with no significant trends in the central and northern Rockies, and advances of 4−13 days for maximum SWE and 9−20 days for complete melting in the southern Rockies. Most of the trends toward earlier melt can be explained by decreases in springtime snowfall (Fig. 8b), and to a lesser extent, by increases in springtime air temperatures and dust deposition. A multiple linear regression model for SM50 using monthly snowfall and air temperatures, and Ca^{2+} concentrations in snowpack and in wet deposition, explained 81% of the variance in SM50 (Fig. 8c). Snowfall (especially in April and March) has the greatest influence (Fig. 8d), causing delays in snowmelt due
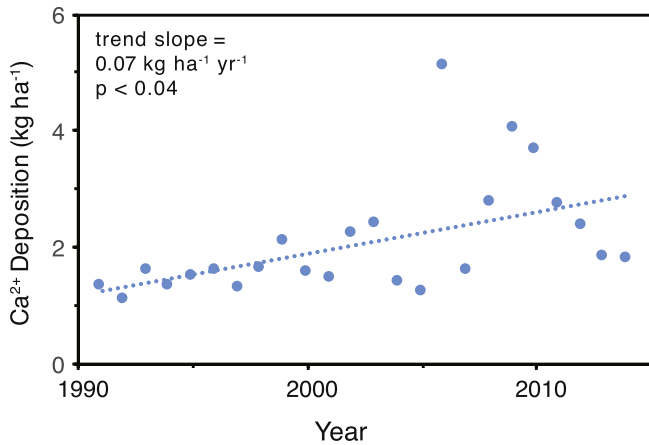
Fig. 6. Annual Ca^{2+} deposition in wet deposition at Loch Vale, Colorado. Dashed line indicates linear regression through data. The trend in Ca^{2+} deposition is driven by changes in concentration, as there is no correlation between annual Ca^{2+} deposition and precipitation amount ($r^2 = 0.001$). Data are from National Trends Network (NTN).
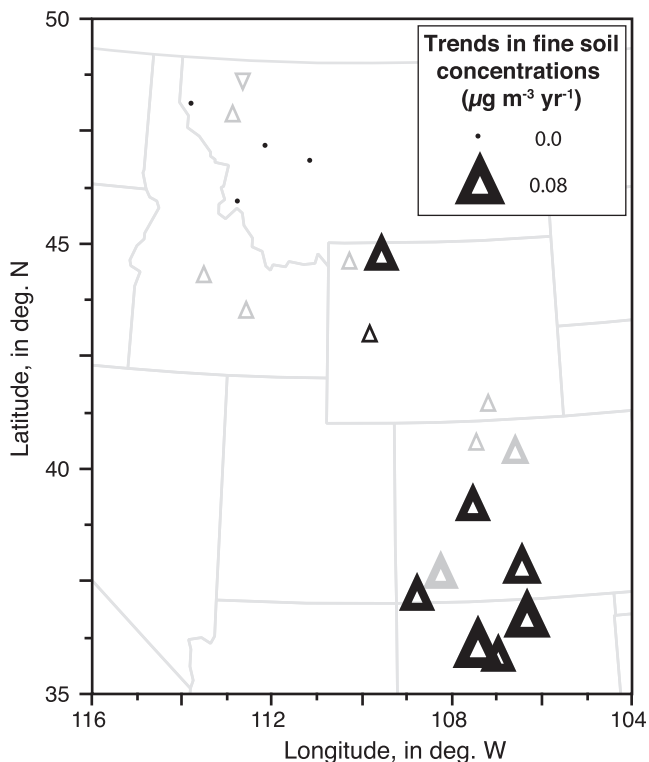

Fig. 7. March trends in fine soil dust particle concentrations in IMPROVE ambient air samples, 1993–2014. Symbols as in Fig. 4 and 5.

to added cold content and because new snow increases albedo by covering old, coarse grained snow and dark snow layers containing dust. Warm air temperatures (especially in May) and dust deposition tend to accelerate snowmelt through increased sensible heat and radiative forcing effects (Fig. 8d).

We investigated the influence of dust deposition further by removing dust surrogate parameters (Ca^{2+} concentrations) from the model, and then plotting the residuals for sites in Colorado during two high-dust years, 2009 and 2013 (Fig. 8e). A negative bias in residuals for these high dust-deposition sites and years would support the hypothesis that dust causes snowmelt to occur earlier

than can be explained by climate variables alone. The median bias in residuals for sites in Colorado was −1.5 days (8.8 to −32.0 days; 95% confidence interval (CI)) for 2009 and −2.4 days (8.2 to −16.9 days; 95% CI) for 2013. At Red Mountain Pass, an area of relatively high dust deposition in southwest Colorado (Skiles et al., 2015), residuals were −6.8 days in 2009 and -8.2 days in 2013. This result provides an indication of the magnitude of the dust radiative forcing effect expected for high dust-deposition scenarios compared to recent average deposition. A regression through the residuals for 2009 and 2013 indicates that bias was greatest in cases of late snowmelt (Fig. 8e). This finding is consistent with those of Painter et al. (2010), who noted that the greatest radiative forcing effects of dust should occur in areas with the deepest snow because they take the longest to melt out.

In an earlier study using a similar approach, Bryant et al. (2013) compared dust radiative forcing in the upper Colorado River Basin to residuals in predicted snowmelt runoff center of mass (COM) from SNOW-17, a process-based snowmelt model. Results indicated a significant negative correlation between dust forcing and COM ($r^2 = 0.42$, $p \leq 0.01$), with observed COM occurring 1.5 ± 0.6 days earlier than predicted for each 10 W m^{-2} of dust forcing. Annual dust forcing during 2000−2010 ranged from 20 to 80 W m^{-2} (Bryant et al., 2013), suggesting that dust accelerated snowmelt by 3−12 days during that period, similar in magnitude to estimates from our empirical model. It should be noted that both models were developed using data covering a range of recent dust deposition and climate conditions (1993−2014 for our model, and 2000−2010 for Bryant et al., 2013). Thus, our estimates of acceleration of snowmelt due to dust deposition are relative to recent average conditions, which may or may not reflect conditions prior to large-scale soil disturbance from grazing. Much larger estimates of snowmelt acceleration due to dust forcing have been reported for high-dust deposition years, such as 2009 or 2013, compared to "clean" snow, containing little dust (Painter et al., 2012; Skiles et al., 2015).

3.6. Implications for snowpack water resources

Anticipated increases in air temperatures and dust deposition to snow may act in concert to dramatically shift the timing of snowmelt in mid-latitude basins where snowmelt is a vital water resource. Mankin et al. (2015) identified 97 basins in the northern Hemisphere where snowmelt provides water during summer and fall to meet demand in excess of that provided by rainfall. These snow-sensitive basins are currently home to ~1.5 billion people (Mankin et al., 2015). Using an ensemble of climate models, they evaluated the likely effect of climate change on snowpack resources in those areas. Results indicated that 10−27 million people live in basins where it is likely that snowmelt will no longer be able to provide sufficient runoff to meet summer/fall demand by 2060. Most of these basins are in the western U.S., southern Europe, and the Middle East, where air temperature increases of 1−3 °C are projected by the mid-21st century (Stocker et al., 2013).

Skiles et al. (2012) calculated radiative forcing effects for anticipated increases in air temperature and from aeolian dust deposition to snow in southwestern Colorado. They estimated that a +2 °C to +4 °C increase in melt-season temperatures at an alpine site would cause a radiative forcing effect of 8−16 W m^{-2}. Estimated radiative forcing from dust deposition to snow ranged from 33 W m^{-2} during a low-dust deposition year (2005) to 50 W m^{-2} during a high-dust deposition year (2009), a difference of 17 W m^{-2}. Thus, during high-dust deposition years, additional radiative forcing from dust is comparable to that expected from increasing air temperatures. Southwestern Colorado receives relatively high dust deposition due to its proximity to dust sources on
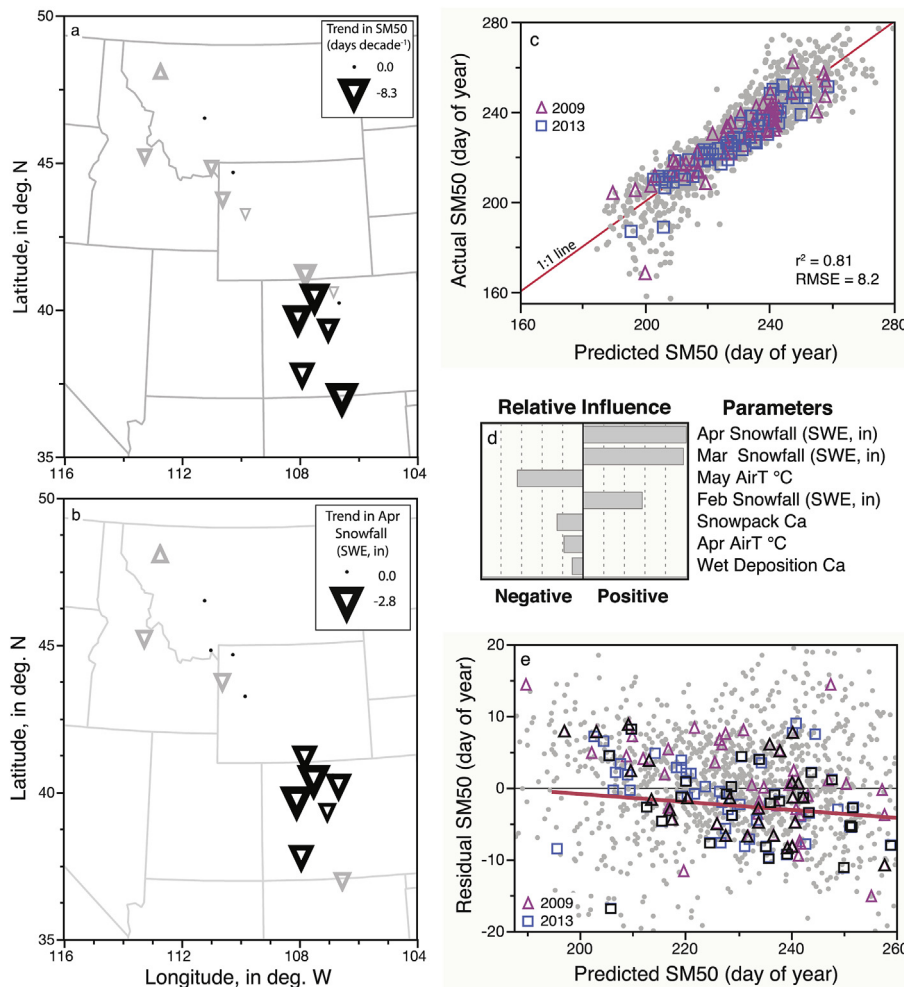
Fig. 8. Maps showing (a) trends in SM50 (day when half of snowpack has melted) at SNOTEL sites, and (b) trends in April snowfall at SNOTEL sites in inches of snow water equivalent (SWE); (c) scatter plot showing actual and predicted SM50 using full multiple linear regression model (MLR), with data from 2009 to 2013 (high dust-deposition years) shown as triangles and squares, and data from other years (1994–2014) as grey dots, (d) relative influence of full MLR parameters, and (e) scatter plot showing residuals from MLR using only climate parameters, with symbols as in 8c, except Colorado sites from 2009 to 2013 in black. In (a) and (b), symbols as in Fig. 4 and 5. In (e), red line shows regression through 2009 and 2013 residuals for Colorado sites; slope of regression is − 0.06.

the Colorado Plateau, and thus the dust effect on snowmelt timing is relatively high as well. Nonetheless, previous studies have indicated that aeolian dust deposition to snow may be important in other mid-latitude basins in southern Europe and Asia (Delmas et al., 1996; Dong et al., 2009). Paleorecords suggest a doubling of aeolian dust emissions during the 20th century across much of the globe, with drought being a major driver (Mahowald et al., 2010). Despite the uncertainty of the paleorecords, it is clear that deposition of aeolian dust to snow poses a substantial risk to snowpack water resources in mid-latitude basins, and the phenomenon is of more than just local concern.

While paleorecords can provide information on long-term changes in dust deposition, sites are too sparse to permit analyses of spatial patterns at high resolution. There are no long-term (e.g., >20 years), regional networks for monitoring dust deposition to snow, so it is necessary to use indirect data from a combination of sources to infer trends in dust deposition (Miller et al., 2004). In this study, we showed how chemistry data from snowpack, wet deposition, and aerosol monitoring networks can be used to evaluate spatial patterns and temporal trends in dust deposition to snow at high resolution. Inferred dust deposition increased from north to south in the Rocky Mountains, reflecting differences in proximity to

dust sources in the southwestern U.S. Calcium concentrations in snow increased by 44%, 65%, and 81% in the northern, central, and southern Rockies, and alkalinity showed similar patterns, indicating that winter/spring dust deposition increased substantially between 1993 and 2014. Snowmelt timing shifted 7–18 days earlier in the southern Rockies over the same time period, most likely due to decreasing snowfall and increasing dust deposition during spring. Climate forecasts for increased frequency and severity of drought in snow-sensitive, mid-latitude basins in the northern Hemisphere suggest that dust deposition to snow will remain an issue of concern for the foreseeable future. The methods used in this study have broad applicability, and similar analyses could be performed in other locations where suitable precipitation and aerosol chemistry monitoring data exist.

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Appendix A. Supplementary data

Supplementary data related to this article can be found at http://dx.doi.org/10.1016/j.atmosenv.2016.06.076.

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