Impacts of anthropogenic emissions and cold air pools on urban to montane gradients of snowpack ion concentrations in the Wasatch Mountains, Utah

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Abstract

Urban montane valleys are often characterized by periodic wintertime temperature inversions (cold air pools) that increase atmospheric particulate matter concentrations, potentially stimulating the deposition of major ions to these snow-covered ecosystems. We assessed spatial and temporal patterns of ion concentrations in snow across urban to montane gradients in Salt Lake City, Utah, USA, and the adjacent Wasatch Mountains during January 2011, a period of several persistent cold air pools. Ion concentrations in fresh snow samples were greatest in urban sites, and were lower by factors of 4 – 130 in a remote high-elevation montane site. Adjacent undeveloped canyons experienced significant incursions of particulate-rich urban air during stable atmospheric conditions, where snow ion concentrations were lower but not significantly different from urban sites. Surface snow ion concentrations on elevation transects in and adjacent to Salt Lake City varied with temporal and spatial trends in aerosol concentrations, increasing following exposure to particulate-rich air as cold air pools developed, and peaking at intermediate elevations (1500 – 1600 m above sea level, or 200 – 300 m above the valley floor). Elevation trends in ion concentrations, especially NH_4^+ and NO_3^-, corresponded with patterns of aerosol exposure inferred from laser ceilometer data, suggesting that high particulate matter concentrations stimulated fog or dry ion deposition to snow-covered surfaces at the top of the cold air pools. Fog/dry deposition inputs were similar to wet deposition at mid-elevation montane sites, but appeared negligible at lower and higher-elevation sites. Overall, snow ion concentrations in our urban and adjacent montane sites exceeded many values reported from urban precipitation in North America, and greatly exceeded those reported for remote snowpacks. Sodium, Cl^-, NH_4^+, and NO_3^- concentrations in fresh snow were high relative to previously measured urban precipitation, with means of 120, 117, 42, and 39 $\mu eq\ l^{-1}$,

respectively. After exposure to atmospheric particulate matter during cold pool events, surface snow concentrations peaked at 2500, 3600, 93, and 90 μeq l^{-1} for these ions. Median nitrogen (N) deposition in fresh urban snow samples measured 0.8 kg N ha^{-1} during January 2011, with similar fog/dry deposition inputs at mid-elevation montane sites. Wintertime anthropogenic air pollution represents a significant source of ions to snow-covered ecosystems proximate to urban montane areas, with important implications for ecosystem function.

Key words: inversion, montane, nitrogen deposition, PM2.5, snow chemistry, urban

1. Introduction

Snow represents the dominant form of precipitation in many arid montane regions and provides a primary source of nutrient ions to ecosystems, with potentially detrimental impacts on water quality and ecosystem function when ion inputs are enhanced by anthropogenic emissions (Lewis et al. 1983, Cerling and Alexander 1987, Jeffries 1990, Williams and Tonnessen 2000). In particular, nitrogen (N) inputs from atmospheric deposition can contribute to undesired increases in productivity, species changes, and aquatic eutrophication, especially in seasonally snow-covered environments where nutrients rapidly elute during melting (Jeffries 1990, Williams and Tonnessen 2000, Baron et al. 2011). Many studies of snow chemical composition over the past three decades have explicitly avoided measurements of urban emission hot-spots (Jeffries 1990, Nickus et al. 1997), instead focusing on remote wilderness sites (Williams and Melack 1991, Pomeroy et al. 1999, Kang et al. 2004, Williams et al. 2009). Cities often exhibit higher ion concentrations and fluxes in rainfall and dry deposition relative to adjacent areas, with trends varying among chemical species (e.g. Lewis et al. 1984, Gatz 1991, Fenn et al. 2003).

However, urban ion deposition studies have typically focused on summer rainfall (Lovett et al. 2000, Bettez and Groffman 2013, Rao et al. in press), and very few comprehensive measurements of snow ions have been reported in urban environments (Lewis et al. 1983). Little is known about spatial and temporal variation in ion deposition to snow across urban to rural gradients, and whether snow ion concentrations vary systematically with urban proximity as observed for rainfall. Even less is known about temporal couplings between urban air pollution events and ion inputs to snow. These patterns may be critical for understanding and predicting nutrient inputs to seasonally snow-covered ecosystems.

Wintertime meteorological characteristics of urbanized montane valleys have the potential to significantly increase snowpack ion loading in wet, dry, and cloud deposition. High pressure events persisting for days to weeks often stabilize cold air pools, trapping local anthropogenic emissions, and leading to the accumulation of ion-rich primary and secondary fine particulate matter (PM 2.5) in the atmosphere (Silcox et al. 2012, Kelly et al. 2013, Lareau et al. 2013, Whiteman et al., 2014). These conditions prevail in many rapidly growing metropolitan areas in the Western United States (e.g. Salt Lake City, Utah; Boise, Idaho; Reno, Nevada) and physiographically similar regions worldwide (e.g. Santiago, Chile; Chen et al. 2012, Cereceda-Balic et al. 2012). Cold air pools associated with elevated PM 2.5 also occur in non-mountainous regions over shorter timescales (Wallace and Kanaroglou 2009), and could thus represent a widely important phenomenon for wintertime urban ion deposition.

Empirical studies linking atmospheric aerosol concentrations with ion deposition remain uncommon, especially for snow (Yalcin et al. 2006, Dolislager et al. 2012). Ammonium nitrate and ammonium sulfate originating from anthropogenic precursors—ammonia, nitrogen oxides, and sulfur dioxide—comprise the majority of PM 2.5 in many urban areas (Connell et al. 2006,

Hand et al. 2012, Kelly et al. 2013). The impact of urban atmospheric emissions and particulate matter on snowpack ion loading is largely unknown, but Cerling and Alexander (1987) documented extremely high concentrations of ions in rime and snow at sites in and adjacent to Salt Lake City during a prolonged cold air pool in 1985. Dry and fog deposition of gases and particles to snow-covered surfaces have previously been shown to comprise relatively minor sources of ions in comparison with snowfall (Bergin et al. 1995, Björkman et al., 2013). Rather, net losses of NO_3^- from the snowpack between precipitation events can be significant (Williams and Melack 1991, Pomeroy et al. 1999, Williams et al. 2009). However, this subject has received little attention in urban environments, where elevated aerosol concentrations during cold air pools could enhance dry or cloud deposition in addition to wet deposition, and yield ecologically significant fluxes of ions to the snowpack.

Here, we assessed spatial and temporal patterns of ion concentrations in bulk snow and snow surface samples across urban to montane gradients in Salt Lake City, Utah, USA, and the adjacent Wasatch Mountains. This major metropolitan area experiences frequent wintertime atmospheric temperature inversions that promote particulate matter accumulation over time, yielding PM 2.5 concentrations that routinely violate national ambient air quality standards (Silcox et al. 2012, Kelly et al. 2013, Whiteman et al. 2014). We demonstrated that urban air pollution associated with cold air pools significantly affected spatial and temporal patterns of snow ion concentrations, with implications for water quality and ecosystem function.

2. Methods

2.1 Sampling locations

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We sampled ion concentrations of snow collected in the Salt Lake City, UT, USA metropolitan area and the adjacent Wasatch Mountains (Fig. 1) during December 2010 – January 2011, including multiple persistent cold air pools (PCAPS) described in an intensive atmospheric observational study by the same name (Lareau et al. 2013). Sampling locations spanned urban Salt Lake City and adjacent montane ecosystems to assess variation in snow chemical composition with landscape position, elevation, and time. We collected two distinct types of samples: bulk snow and surface snow. Sampling elevations are expressed in meters above sea level; the valley floor has an approximate elevation of 1300 m.

2.2 Bulk snow samples

We sampled fresh bulk snow samples at three urban sites and along two elevation gradients in adjacent montane canyons (Fig. 1). Urban sites comprised a roof on the University of Utah campus, a residential yard, and an elementary school. The two montane canyons, Red Butte and Big Cottonwood, experienced elevated particulate matter concentrations during cold air pools (Fig. 2). Red Butte Canyon is a Research Natural Area managed by the USDA Forest Service with restricted access and minimal vehicle traffic. Big Cottonwood Canyon contains a highway and two major ski resorts. We sampled each canyon at elevations of approximately1600, 1800, and 2000 m, respectively. We also sampled an additional very high elevation site (2900 m) above Big Cottonwood Canyon. Sites were sampled 5 – 7 times between December 17, 2010 and January 30, 2011 (total n = 51 bulk snow samples).

Bulk snow samples were collected on rigid high-density polyethylene (HDPE) sampling surfaces mounted on white plywood (0.6 x 0.6 m) with a 2 m vertical pole (hereafter termed "stormboards", n = 10), deployed in the field for the entire study period. Stormboards were located in undisturbed clearings and sampled within 24 – 48 hours after each snowfall by

vertically coring from the top of the snowpack to the HDPE surface with a 5.1 cm diameter PVC tube. Samples were quantitatively transferred to HDPE bottles by placing a plastic knife under the tube. Two samples were collected from each stormboard and composited. We removed remaining snow from stormboards following sampling and rinsed surfaces with deionized water. Stormboards were replaced on top of the snowpack in an adjacent undisturbed location. All sampling supplies were acid-washed and rinsed with deionized water before use, and clean nitrile gloves worn during sample collection. Field blanks were collected to assess contamination by rinsing sampling cores with deionized water in the field. Ion concentrations in field blanks represented < 3 % of mean snow concentrations for all ions except for K^+ (~10 % of sample concentrations). This variation (3 %) only slightly exceeded the analytical uncertainty of ion analyses (assessed using the standard deviation of check standards). We calculated snow water equivalent for bulk snow samples by dividing snow sample mass by sampling area.

2.3 Snow surface samples

Snow surface samples were repeatedly collected from two elevation transects (total n = 90 snow surface samples; Figs. 1 and 2). The urban transect was located on a south-facing slope in the Avenues neighborhood of Salt Lake City at the same seven sites described by Silcox et al. (2012). The montane transect was located on a west-facing ridge of Grandeur Peak, a mountain immediately East of Salt Lake City. This transect comprised 20 sampling sites located every 50 meters in elevation from 1550 to 2500 m. Snow surface samples were collected from each site by scraping the mouth of a 60 ml HDPE bottle across the snow surface to a depth of approximately 1 cm. The montane and urban transects were sampled four and five times, respectively, during January 2011.

2.4 Chemical analyses

Thawed samples were vacuum filtered through pre-rinsed Whatman Nuclepore 1.0 μm membranes and analyzed for major anions (Cl^{-1}, SO_4^{2-}, NO_3^-) by ion chromatography and cations (Na^+, Ca^{2+}, NH_4^+, Mg^{2+}, K^+) by atomic absorption spectrometry at the Kiowa Environmental Chemistry Laboratory at the University of Colorado, as described in Williams et al. (2009). Detection limits for all analytes reported here were < 0.5 μeq l^{-1}, with analytical precision < 2 %. A subset of samples (n = 33) was also analyzed for acid neutralizing capacity (ANC) and pH to calculate charge balance (carbonate species were assessed indirectly via ANC). Most samples had a small excess positive charge with a median ion percent difference (difference of anion charge and cation charge divided by their total) of -8 %. Unmeasured anions including organic acids, nitrite, and fluoride likely accounted for this discrepancy.

2.5 Meteorological and air-quality measurements

Concentrations of PM 2.5 were measured hourly by the Utah Department of Environmental Quality at a site co-located with the lowest elevation on our urban transect (Figure 1; data available at http://www.airmonitoring.utah.gov/dataarchive/archpm25.htm). We compared temporal relationships between PM 2.5 concentrations and ion concentrations in surface snow samples. For each sample, we integrated hourly PM 2.5 concentrations from the previous snowstorm until the time of sampling as a metric of potential exposure to PM2.5, irrespective of elevation. Snow accumulation at the mouth of Little Cottonwood Canyon was recorded daily. Near-surface air temperatures were measured along a pseudo-vertical transect (50 m elevation contours) on the montane transect with sensors (Hobo Pro, Onset Computer Corporation, MA) mounted on fence posts at ~1.2 m height; we report mean temperature profiles from several days to illustrate contrasting atmospheric conditions during cold air pool development.

2.6 Aerosol exposure index

We recorded aerosol backscatter profiles with a Vaisala CL31 laser ceilometer located at the center of the Salt Lake Basin (Fig. 1). An index for exposure of the surface snow to aerosol loading from the adjacent atmosphere was derived as the time integral of the backscatter coefficient β between the last snowfall and the sampling time. As the log10 of β yields negative numbers and -8 can be considered the minimum value for log10 β, we added 8.0 to backscatter data to achieve a positive scale for integrated exposure. This metric implies that higher exposure occurs at elevations where the atmosphere exhibited the largest values of aerosol backscatter for the longest time. Backscatter represents a semi-quantitative measure of aerosol abundance that is not directly proportional to concentrations of a single aerosol species, as backscatter depends on particle size and composition. Both the backscatter coefficient in Fig. 5 and the aerosol exposure index (Fig. 7) thus represent semi-quantitative approximations of aerosol concentrations or exposure.

2.7 Statistical analysis

We used ANOVA and linear mixed effects models to evaluate spatial and temporal differences in ion concentrations among sites and dates. Ion concentration data were log-transformed to reduce heteroscedasticity. For bulk snow samples, we compared ion concentrations among sites over the entire sampling period using ANOVA. For surface snow samples, we fit two separate models for each ion. First, we assessed differences among sampling dates irrespective of elevation, including random effects for sites to account for repeated sampling. Pairwise differences among dates were evaluated with Tukey comparisons. Secondly, we fit models including sampling date, elevation, and elevation x date interactions as fixed effects, and sampling sites as random effects. Ion concentrations displayed strong nonlinear

trends with elevation for one sampling date (January 28) in the urban transect, so we fit regression models including a quadratic term for this date. We fit all models using the lm or lme functions in R (Pinheiro et al. 2013).

3. Results and Discussion

3.1 Bulk snow ion concentrations

Concentrations of major ions varied by up to four orders of magnitude among bulk stormboard snow samples and showed significant spatial variation from urban to montane sites. Sodium and Cl^{-1} had the highest concentrations in bulk and surface snow samples, followed by Ca^{2+}, NH_4^+, NO_3^-, SO_4^{2-}, Mg^{2+}, and K^+, respectively (Fig. 3). Ion concentrations in bulk snow samples decreased significantly with elevation from the urban sites to the very high elevation Big Cottonwood Canyon site for all species except for K^+, whereas sites in Big Cottonwood and Red Butte Canyons were typically intermediate and not significantly different. (Fig. 3). Mean concentrations of ions in the urban samples relative to the very high elevation site were greater by factors of 130 (Cl^{-1}), 31 (Na^+), 8 (Ca^{2+}), 12 (NH_4^+), 8 (NO_3^-), 8 (SO_4^{2-}), 11 (Mg^{2+}), and 4 (K^+). Proton concentrations did not differ among sites (7.4 ± 1.1 μeq l^{-1}, $n = 40$), and were equivalent to pH 5.13 ± 0.07.

Despite variation in ion concentrations between the urban and canyon sites, ion mass loading appeared similar across the landscape due to weak inverse relationships between snow water equivalent and ion concentrations (Fig. A1 and A2). These patterns suggest that increasing snow water equivalent with elevation due to orographic precipitation may be as important as spatial variation in ion concentrations in controlling ion mass fluxes. However, we note that estimates of ion mass loading, as opposed to ion concentrations, are inherently imprecise in

montane environments due to spatial heterogeneity in snow water equivalent, which our study was not designed to assess (Molotch and Bales 2005). Furthermore, snow ion concentrations are especially relevant to biological processes in open systems due to the fundamental dependence of nutrient uptake rates and osmotic relations on ion concentration gradients, as opposed to ion mass fluxes.

3.2 Pairwise correlations among major ions

Almost all ion pairs exhibited significant positive correlations except for H^+ (Table 2), indicating coherence in distinct sources of these ions. Notably, correlations among ions tended to be substantially weaker in the surface snow samples relative to the bulk snow samples, suggesting a mixture of different sources and/or depositional processes (wet + dry + fog). One exception was the strong correlation between Ca^{2+} and SO_4^{2-} in surface snow, likely reflecting gypsum dust from nearby salt flats. Sodium and Cl^- had the strongest possible correlation ($r = 1$) in both the bulk and surface snow samples, strongly implicating NaCl aerosol inputs. The next strongest correlations were between NH_4^+, SO_4^{2-}, and NO_3^-, likely reflecting inputs of particulate $(NH_4)_2SO_4$ and NH_4NO_3, which frequently represent dominant constituents of PM 2.5 in this region and nationwide (Hand et al. 2012, Kelly et al. 2013). Calcium and Mg^{2+} (but not K^+) also scaled closely, potentially indicating a common source in carbonate dust or ash (Lajtha and Jones, 2013).

3.3 Comparisons of snow ion concentrations with previous studies

Concentrations of NH_4^+, NO_3^-, Na^+, and Cl^- in the urban samples were several times greater than previous measurements in snow from regions with similar climate and physiography (Table 1). Sodium and Cl^- were the dominant ions across all sites. The high concentrations of Na^+ and Cl^- (tens – hundreds of $\mu eq\ l^{-1}$) we observed in our urban sites were similar to those

documented in snow in Montreal, Canada, which were likely derived from road deicing salt aerosols (Lewis et al. 1983). The highly saline margins of the Great Salt Lake may contribute to high background Na^+ and Cl^- evident in the canyon and remote sites compared with other montane snow studies (Table 1). Our urban bulk snow Na^+ and Cl^- concentrations were comparable to mean precipitation measurements in maritime regions such as Athens, Greece (~90 μeq l^{-1}), and slightly greater than Los Angeles, Californita (~45 μeq l^{-1}; Gatz 1991), suggesting high background inputs from lake-derived aerosols in addition to episodic road salt additions. Anthropogenic sources likely contributed to the sporadic occurrence of extremely high NaCl at some sites (discussed below).

Concentrations of NH_4^+ and NO_3^- in Salt Lake City snow were generally greater than those in rainfall from other North American cities, and from one other study that measured urban snow ion concentrations. We acknowledge that substantial spatial and temporal variability exists within cities and among calibrations and analytical methods used in the studies reported here, but we provide mean values from other studies to facilitate general comparisons, irrespective of their possible statistical significance. We found four-fold greater concentrations of NH_4^+, similar NO_3^-, and much lower SO_4^{2-} as compared with snow from urban Montreal measured in the early 1980's (Table 1; Lewis et al. 1983). Our urban bulk snow typically had greater NH_4^+ concentrations than mean wet precipitation measured in Detroit, Chicago, Los Angeles, and New York City in the 1980's (approximately 26, 35, 20, and 15 μeq l^{-1} for each city, respectively), as well as greater NO_3^- concentrations than these cities (approximately 36, 32, 31, and 27 μeq l^{-1}, respectively; Gatz 1991). Urban snow NO_3^- concentrations were similar to those of bulk summer rain precipitation measured in downtown New York City in 1996 (~40 μeq l^{-1}), but our NH_4^+ concentrations were substantially greater than the 25 μeq l^{-1} measured there (Lovett et al. 2000).

Our urban snow also had greater NH_4^+ and NO_3^- concentrations compared with most recent precipitation samples (2008 – 2010, rain and snow) in National Atmospheric Deposition Program (NADP) sites documented in a meta-analysis, which typically measured < 25 μeq l^{-1} (Lajtha and Jones 2013). These sites were explicitly located to minimize the direct influence of urban sources. Salt Lake City snow NH_4^+ and NO_3^- concentrations were similar to several European Monitoring and Evaluation Programme sites reported in this study (ibid.).

3.4 Air quality dynamics during the snow sampling period

Daily mean PM 2.5 concentrations at the Salt Lake Valley floor varied between 10 – 70 μg m^{-3} during January 2010 (Fig. 4) concomitant with cold air pool dynamics (Lareau et al. 2013). Three distinct cold air pool episodes of Jan 1 – 9, 12 – 16, and 26 – 31 are visible in Fig. 4 as periods of increased PM 2.5 surface concentrations. Aerosol backscatter increased during these cold air pool episodes when mixing was inhibited by stable stratification in the atmosphere (Fig. 5). The height of the aerosol layer varied in response to changing meteorological conditions, with the top typically positioned between 1500 – 2000 masl. When fog or a low stratus cloud deck formed, the ceilometer laser beam was occasionally completely attenuated. This led to signal saturation and elimination of returns above the fog/cloud layer during the nights between January 6 – 8, and between January 27 – 30.

3.5 Temporal variation in surface snow ion concentrations

Surface snow samples from the urban and montane elevation transects exhibited significant temporal variation in ion concentrations approximately proportional to cold air pool development and time-integrated PM 2.5 concentrations measured on the valley floor (Fig. 4). Irrespective of elevation, ion concentrations in the montane transect were significantly greater on either Jan 7, or both Jan 7 and 30 as compared with Jan 14 and 24 for several ions (Cl^-, Na^+,

Ca^{2+}, K^{+}; Fig. 4). The dates with significantly greater snow surface ion concentrations coincided with periods of greater time-integrated PM 2.5 exposure, calculated by multiplying valley surface PM 2.5 concentrations by time between snow events (indicated by the size of shaded regions in Fig. 4). On Jan 7 and 30, time-integrated PM 2.5 exposure reached 4760 and 3070 μg m^{-3} hr, whereas the dates with significantly lower surface ion concentrations (Jan 14 and 24) had much lower PM 2.5 exposure (930 and 300 μg m^{-3} hr). In the urban transect, concentrations of Ca^{2+}, NO_3^{-}, and SO_4^{2-} were significantly greater on Jan 28 than on the other sampling dates, coinciding with a PM 2.5 exposure of 1340 μg m^{-3} hr. The other sampling dates (Jan 5, 14, 24, and 26) had lower PM 2.5 exposures of 1090, 930, 300, and 350 μg m^{-3} hr, respectively.

The potential influence of road salt on surface snow Na^{+} and Cl^{-} was especially evident in the montane samples on Jan 7, when concentrations increased by an order of magnitude to thousands of μeq^{-1} between 1600 – 1800 m (Fig. 6). High deposition of Na^{+} and Cl^{-} to the montane transect could have been generated by a combination of road salt application and high-speed vehicle traffic on Interstate Highways 80 and 215, which are located within several hundred meters of the lower sites (Fig. 1); the urban elevation transect was not located with similar proximity to major highways.

3.6 Variation in surface snow ion concentrations and particulate matter with elevation

Elevational trends in surface snow ion concentrations differed between the urban and montane transects (Fig. 6). On the montane transect, ion concentrations consistently decreased with elevation ($p < 0.0001$) by a relationship that varied among sampling dates ($p < 0.0001$, elevation x date interaction). Samples collected on January 7 and 30, during cold air pools with greatest PM 2.5 exposure, exhibited the greatest elevational differences along the transect (Fig. 6). In the urban transect, significant linear trends were evident only for Na^{+} and Cl^{-}, which

decreased with increasing elevation ($p < 0.05$ for elevation and elevation x date interaction). However, several ions in the urban transect exhibited increasingly nonlinear concentration trends with elevation during the development of a prominent temperature inversion between January 24 – 30 (Figs. 5-7). On January 24 and 26, the daily mean temperature profile showed a temperature decrease with height approximately following a lapse rate of 5.5 °C km^{-1}, corresponding with a nearly uniform elevational aerosol exposure profile indicated by ceilometer backscatter (Fig. 7). During these dates, surface snow NH_4^+ and NO_3^- concentrations varied little with elevation, although a subtle trend of increasing NH_4^+ with decreasing elevation was evident in both transects. However, by January 28 and 30, a cold pool developed with a strong elevated temperature inversion layer between 1600 – 2000 masl. Peak aerosol exposure occurred between 1500 – 1600 masl corresponding with the vertical structure of this cold air pool (Fig. 7). The two other cold air pools during January 2011 showed similar trends, although exposure peaked at slightly lower elevations (1400 – 1500 m; Fig. A3). Corresponding with increased aerosol exposure, concentrations of NH_4^+ began to increase at lower elevations on January 26, and by January 28, NH_4^+ and NO_3^- significantly peaked at approximately 1500 masl in the urban transect (Fig. 7; $p < 0.01$ and $p < 0.05$ for quadratic relationships with elevation, respectively). Similarly, by January 30, concentrations of these ions peaked in the montane transect at approximately 1600 masl.

Non-linear elevational trends in aerosol exposure and ion accumulation during cold air pool development are generally consistent with elevation trends in PM 2.5 concentrations measured by Silcox et al. (2012) on the montane transect, which peaked between 1350 – 1400 m on January 6 and 28. Subtle differences between elevational trends in PM 2.5 and our exposure index (Fig. 7), which peaked at slightly higher elevations, may have been due to the fact that the

ceilometer measured aerosols regardless of their diameter, whereas PM 2.5 measurements by definition are restricted to a fine subset of particulate matter (< 2.5 μm). These differences could also have been influenced by horizontal spatial variability in aerosol concentrations between the location of the ceilometer and the urban transect, although PM 2.5 concentrations measured at several sites within the Salt Lake Valley were very similar during Jan 2011 (Silcox et al. 2012). The improved correspondence between elevation trends in surface snow ions and our index of aerosol exposure, as opposed to elevation trends in PM 2.5, suggests that coarser aerosols (> 2.5 μm) might represent an important source of ions to snow.

3.7 Nitrogen deposition fluxes

We cannot conclusively separate fog, hoar, rime, and/or dry deposition ion inputs (collectively termed "fog/dry deposition") from wet snowfall, but can estimate their relative importance. Fresh bulk snow likely contains mostly wet ion deposition, but may also capture some fog/dry inputs deposited to the stormboard surface prior to snowfall. Bulk snow samples provide maximum estimates of wet deposition, whereas comparison of surface snow samples over time yields potential net fog/dry deposition. As a cold air pool developed between Jan 24 and Jan 30, changes in surface snow NH_4^+ concentrations over time across the montane gradient varied between a decrease of 11 μeq l^{-1} and a maximum increase of 62 μeq l^{-1} at mid elevations (Fig. 7). Nitrate showed similar patterns, where concentrations increased between 0 and 70 μeq l^{-1} over this period. The urban transect showed similar increases between Jan 24 and Jan 28 of 15 – 38 and 29 – 56 μeq l^{-1} for NH_4^+ and NO_3^-, respectively. These trends reflect significant net positive fog/dry N deposition at mid-elevation sites, and negligible deposition or small net N losses at the low and high-elevation sites.

The highest rates of surface deposition in the montane and urban transects were equivalent to N fluxes of approximately 0.08 – 0.10 kg N ha^{-1} for each species (NH_4^+ and NO_3^-) over these four and six-day periods. Maximum rates of fog/dry deposition at mid-elevation sites during the late January cold air pool were comparable to deposition from individual storm events (bulk snow samples) in the urban area, which had median values of 0.06 - 0.05 kg N ha^{-1} for NH_4^+ and NO_3^- (Fig. A2). The seven measured snow events in January 2011 deposited a total median N input of approximately 0.8 kg ha^{-1}, with a smaller additional contribution from fog/dry deposition that varied strongly with elevation. At mid-elevation (1400 – 1600 m) sites, fog/dry deposition may have deposited approximately 0.5 kg N ha^{-1} over the three January cold air pools, if rates of snow surface N accumulation from January 24 - 30 are representative of the earlier cold air pools. At elevations less than 1400 and greater than 1800 m, however, net fog/dry deposition appeared to be much smaller.

3.8 Implications for ecosystem function

Baron et al. (2011) showed that N loading of several kg ha^{-1} yr^{-1} can represent an ecological critical load for nutrient enrichment in Western US montane ecosystems. Given that our median measured N deposition measured almost 1 kg ha^{-1} in January 2011 alone, wintertime deposition can represent an ecologically relevant source of N to these urban and protected montane sites. Summertime N deposition remains to be measured in this region. If our data are representative of other cities, they have significant implications for quantifying N deposition to other urban ecosystems, given that wintertime data were absent from several previous urban deposition studies (Rao et al. In press, Lovett et al. 2000, Bettez and Groffman 2013). Wintertime N inputs have the potential to alter community structure and ecosystem function as

they are delivered to soils and streams during snowmelt (Jeffries 1990, Williams and Tonnessen 2000, Williams et al. 2009, Baron et al. 2011).

Episodically high inputs yielding high aqueous concentrations of Na^+ and Cl^- also have important ecological implications. Some aquatic organisms, including amphibians, are highly sensitive to elevated ion concentrations. Solution conductivities of 500 μS cm^{-1} (equivalent to NaCl concentrations of several hundred μM) can decrease larval survival (Karraker et al. 2008). We found peak NaCl concentrations exceeding 3000 μM in surface snow on the montane transect (Fig. 6), likely derived from road salt aerosols. Snowpack-integrated NaCl concentrations are presumably lower than those found in surface snow, but ion concentrations in initial snowmelt increase as a consequence of preferential ion elution, and are likely to be of similar magnitude to those found detrimental for amphibian survival. Detailed relationships between snowpack and stream ion concentrations are beyond the scope of the present study. The canyons adjacent to the montane elevation transect are "protected" watersheds managed by the USDA Forest Service to maintain a clean drinking water supply for urban use and for conservation purposes. The Wasatch Mountains contain four large federally-designated wilderness areas. Nevertheless, our data show that they experience significant ion pollutant inputs as a consequence of their proximity to the urban Salt Lake Valley. High snow inputs of other ions such as Ca^{2+}, Mg^{2+}, and SO_4^{2-} are unlikely to have substantial ecological effects in this region given the prevalence of sedimentary parent material and high stream concentrations of these ions (S. Hall, unpublished data).

3.9 Couplings between ion deposition and atmospheric particulate matter dynamics

The qualitative correspondence between increased concentrations of major ions in surface snow (Fig. 6), elevational trends in atmospheric particulate matter (Figs. 5 and 7), and

differences in time-integrated PM 2.5 exposures following snowstorms (Fig. 4) all point to the importance of urban particulate matter deposition as a source of ions to snow in this region—a mechanism that has received scant attention in the literature (Cerling and Alexander 1987). Observed variation in surface snow ions between snow events could have been caused by dry deposition or by the precipitation of surface rime or snow grains from moisture originating within the cold air pool. Composition of PM 2.5 is typically dominated by the major ions, especially NH_4^+, SO_4^{2-}, and NO_3^- (Hand et al. 2012), which are readily soluble in melted snow samples. Thus, variation in PM 2.5 concentrations in urban environments may represent an important proximate control on ion deposition to snow, especially at elevations where aerosol clouds are in close contact with the snow surface. The surface snow ion concentrations we observed were greater than those observed elsewhere in bulk snow, although they were substantially lower than previous measurements of rime and surface snow in the Salt Lake Valley (Table 1; Cerling and Alexander, 1987). These differences may reflect the partial success of emissions regulations in decreasing atmospheric particulates over this time period (Whiteman et al. 2014). Low particulate matter concentrations characteristic of remote montane regions may explain the absence of measurable dry ionic snowpack deposition reported in previous studies (Williams and Melack 1991, Pomeroy et al. 1999, Williams et al. 2009).

It is important to note that although our surface snow measurements focused on sites in close proximity to urban Salt Lake City, high concentrations of urban-derived particulate matter extended into significant portions of adjacent montane canyons (Fig. 2). For instance, vertical trends in aerosol distribution (Fig. 5) implied that low and mid-elevation canyon sites (Fig. 1) were exposed to urban aerosols for much of January 2011. Incursions of urban particulate matter (e.g., Fig. 2) may explain the absence of significant differences between bulk snow ion

concentrations in urban and canyon samples; the very high elevation site was situated well above the urban cold air mass (Figs. 1, 3). In fact, the peak in aerosol exposure that we observed at approximately 1600 m concomitant with increased NH_4^+ and NO_3^- concentrations implies that montane sites at intermediate elevation experience elevated dry/fog ion deposition relative to the urban valley floor, perhaps analogous to a "bathtub ring" of accumulated pollution at the top of cold air pools. The correspondence of elevated snow ions to elevations of maximum ceilometer attenuation suggests that aerosol clouds may have generated aerosol-rich rime on the snow surface. The hypothesis suggested by our data, that wintertime ion deposition peaks in montane environments at the top of cold air pools, merits further exploration.

4. Conclusions

We observed high concentrations of major ions in bulk snow and surface samples collected within and adjacent to Salt Lake City, UT, during a period of several cold air pools that generated increased concentrations of anthropogenic particulate matter in January 2011. Concentrations of Na^+, Cl^-, NH_4^+, and NO_3^-, in particular, were typically greater than those measured in other studies of urban precipitation, and concentrations far exceeded typical values recorded in remote montane snowpacks. Bulk snow ion concentrations were greatest in urban sites, but were not significantly different from nearby canyon sites located in protected areas, likely due to incursions of particulate-rich air. Concentrations of Na^+ and Cl^- were high and equivalent to maritime precipitation at all sites, due to the proximity of the Great Salt Lake. However, the impact of road salt application was evident even in this saline environment when concentrations spiked by an order of magnitude on protected montane sites to ~ 3000 $\mu eq\ l^{-1}$. Surface snow ion concentrations tracked temporal and spatial variation in atmospheric particulate concentrations, and were typically greatest at intermediate elevations (1400 – 1600

masl, or 100 – 300 m above the valley floor). Median N loading accompanying snowstorms masured 0.8 kg ha^{-1} in January 2011, with additional contributions from dry/fog deposition that varied strongly with elevation. Wintertime anthropogenic air pollution and road salt application represent ecologically significant sources of nutrients and salts to the snowpack, with important implications for urban and adjacent ecosystems.

Acknowledgements

This project was inspired by S. Arens, whose research on ion deposition to the high elevation snowpack of the Wasatch Mountains provided a baseline for our study. We thank J. Horel, C. D. Whiteman, G. Silcox, and T. Cerling for discussion, D. Reim for snow depth measurements, and W. Brown from NCAR/EOL for providing the ceilometer. Funding was provided by the Funding Incentive Seed Grant Program at the University of Utah and by NSF grant ATM-0938397 (PCAPS study). We thank the Utah Department of Air Quality for providing PM 2.5 data. This research was supported in part by NSF EPSCoR grant EPS 1208732 awarded to Utah State University, as part of the State of Utah Research Infrastructure Improvement Award. Any opinions, findings, and conclusions or recommendations expressed are those of the author(s) and do not necessarily reflect the views of the National Science Foundation.

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574 **Tables**

575 Table 1: Comparison of mean ion concentrations (μeq l^{-1} with standard deviations in parentheses) in snow samples measured in
576 several previous studies in the Western United States. Wasatch samples represent peak snowpack full column concentrations in 2009
577 from 16 high-elevation locations in the Wasatch Mountains, Utah (Arens 2010). Niwot Ridge is an alpine tundra site in the Rocky
578 Mountains near Boulder, Colorado; concentrations represent snowpack from 2006 - 7 (Williams et al. 2009). Emerald Lake is in the
579 Southern Sierra Nevada Mountains in California, samples represent cumulative snowfall from 1985 – 88 (Williams and Melack 1991).
580 Urban samples from Montreal, Canada were collected in 1980 (Lewis et al. 1983). Samples from sites in Salt Lake City represent
581 mean ion concentrations in rime, surface hoar, and surface snow during a persistent cold air pool in 1985-86 (Cerling and Alexander
582 1987). Salt Lake City bulk snow ion concentrations are from the present study.
583

Location	Elevation (m)	Cl^-	NO_3^-	SO_4^{2-}	NH_4^+	K^+	Na^+	Mg^{2+}	Ca^{2+}
Wasatch Mountains (Utah)	2050 – 3050	40.4 (13)	7.5 (1)	20.5 (5.2)	11.6 (2.6)	7.1 (4.7)	43.6 (15.1)	10.3 (3.3)	57.9 (21.2)
Rocky Mountains (Niwot Ridge, Colorado)	3340	1.7	11.5	7.0	4.5	3.6	2.0	4.2	22.6
Sierra Nevada Mountains (Emerald Lake, California)	2800 - 3400	2.8	2.3	2.0	1.7	0.7	1.3	0.6	1.3
Montreal, Canada (1980)	10 - 40	114 (88)	35 (12)	63 (56)	10 (8)	21 (19)	103 (99)	11 (7)	68 (93)
Salt Lake City (1985-6)	1400	1910	1260	1160	1690	60	1690	240	1220

Salt Lake City bulk snow (this study)	1300 - 1400	117 (118)	39 (35)	25 (33)	42 (43)	3 (3)	120 (124)	11 (8)	36 (26)

Table 2: Pairwise (Pearson) correlation coefficients among ion concentrations in bulk snow samples (**a**) and snow surface samples (**b**). Significant correlations ($p < 0.05$) are denoted by bold-face type.

a) Bulk snow samples

	H^+	NH_4^+	Ca^{2+}	Mg^{2+}	Na^+	K^+	Cl^-	NO_3^-	SO_4^{2-}
H^+	1								
NH_4^+	-0.4	1							
Ca^{2+}	-0.5	**0.59**	1						
Mg^{2+}	-0.46	**0.71**	**0.92**	1					
Na^+	-0.21	**0.88**	**0.58**	**0.73**	1				
K^+	-0.2	**0.45**	**0.55**	**0.57**	**0.45**	1			
Cl^-	-0.22	**0.7**	**0.58**	**0.73**	**1**	**0.45**	1		
NO_3^-	-0.29	**0.94**	**0.57**	**0.56**	**0.74**	0.37	**0.58**	1	
SO_4^{2-}	-0.35	**0.92**	**0.66**	**0.7**	**0.86**	0.34	**0.85**	**0.83**	1

b) Snow surface samples

	H^+	NH_4^+	Ca^{2+}	Mg^{2+}	Na^+	K^+	Cl^-	NO_3^-	SO_4^{2-}
H^+	1								
NH_4^+	0.02	1							
Ca^{2+}	-0.78	**0.75**	1						
Mg^{2+}	-0.7	**0.7**	**0.72**	1					
Na^+	-0.33	0.26	**0.32**	**0.69**	1				
K^+	0.07	**0.44**	**0.31**	**0.44**	**0.49**	1			
Cl^-	-0.35	**0.47**	**0.32**	**0.69**	**1**	**0.48**	1		
NO_3^-	0.54	**0.76**	**0.73**	**0.77**	**0.57**	0.27	**0.63**	1	
SO_4^{2-}	0.6	**0.82**	**0.96**	**0.69**	**0.41**	**0.4**	**0.61**	**0.79**	1

Figure captions

Figure 1: Sampling locations in the Salt Lake Valley metropolitan area and adjacent Wasatch Mountains. Panel b) is enlarged from the box in panel a). Diamonds, pluses, and x marks denote stormboards in Red Butte Canyon, Big Cottonwood Canyon, and urban sites, respectively. Squares and circles denote urban and montane elevation transects. The triangle represents the laser ceilometer.

Figure 2: Images of cold air pools and associated high aerosol concentrations in the Salt Lake Valley. At left is a view of the cold-air pool on January 7, 2011, looking from the montane elevation gradient across the urban area. At right is a photo of a cold-air pool on Dec. 18, 2011, penetrating into Mill Creek Canyon in the Wasatch Mountains adjacent to the Salt Lake Valley.

Figure 3:

Ion concentrations in bulk snow samples plotted by sites ranked according to elevation on the ordinate. Boxes represent medians and the interquartile range; data > 1.5 times the range from the box to the whiskers are denoted as outliers (circles). "Urban" denotes samples from three urban locations; low, mid, high, and very-high prefixes represent elevations in Red Butte (RB) and Big Cottonwood (BC) canyons, respectively. Sites with different lowercase letters were significantly different (Tukey comparison, $p < 0.05$). An outlier with a Cl^- concentration of 2140 $\mu eq\ l^{-1}$ in an urban sample was omitted for clarity.

Figure 4: Daily mean concentrations of particulate matter (PM 2.5, black line) and snow surface ion concentrations plotted by date during January 2011 for the urban (red) and montane (blue)

elevation transects. The U. S. EPA national ambient air quality standard is 35 μg m^{-3} (24 hr mean). Grey shaded areas represent time-integrated PM 2.5 exposure of surface snow between the prior snow precipitation event and a given sampling date. Ion concentrations were significantly greater on either January 7, or both January 7 and 30, as compared with January 14 or 24 for several ions (Cl^-, Na^+, Ca^{2+}, K^+) in the montane transect. Concentrations of Ca^{2+}, NO_3^-, and SO_4^{2-} in the urban transect were significantly greater on January 28 than the other sampling dates (January 5, 14, 24, and 26). Temporal trends in Na^+, Mg^{2+}, SO_4^{2-}, and K^+ are shown in Fig A3.

Figure 5: Time-height cross-section of aerosol backscatter (log_{10} of backscatter coefficient β in m^{-1} sr^{-1}), measured with a laser ceilometer. Brighter colors (reds) indicate regions and periods of increased backscatter (more positive β) as indicated in the legend. Precipitation events are indicated by vertical dashed lines with new snow accumulation totals (measured near the mouth of Little Cottonwood Canyon) given on top of the figure. Time intervals between snow precipitation and surface snow sampling events are denoted by blue and red lines below the figure for the montane and urban transects, respectively.

Figure 6: Snow surface ion concentrations by elevation and sampling date during January 2011 for the urban (red) and montane (blue) elevation transects. Different symbols denote different sampling dates. Sodium and Cl^{-1} concentrations were an order of magnitude higher on January 7 than other dates (see insets with distinct scales). Ion concentrations decreased with elevation in the montane transect according to trends that significantly differed among sampling dates. Ion

concentrations generally did not covary with elevation in the urban transects, but NH_4^+ and NO_3^- showed significant non-linear elevational trends on January 28.

Figure 7: Daily average near-surface air temperatures, aerosol exposure index (see Methods), and surface snow NH_4^+ and NO_3^- concentrations as a function of elevation during late January 2011 (dates shown in the legends). The top panels represent two dates with relatively unstable atmospheric conditions, when temperatures decreased at approximately 5.5 °C km^{-1} (dotted lines, left panels), and snow surface ions showed little relationship with elevation. The bottom panels illustrate dates with a strong temperature inversion between 1600 – 2000 masl, leading to increased aerosol exposure and snow surface N concentrations.

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663 Figure 1:

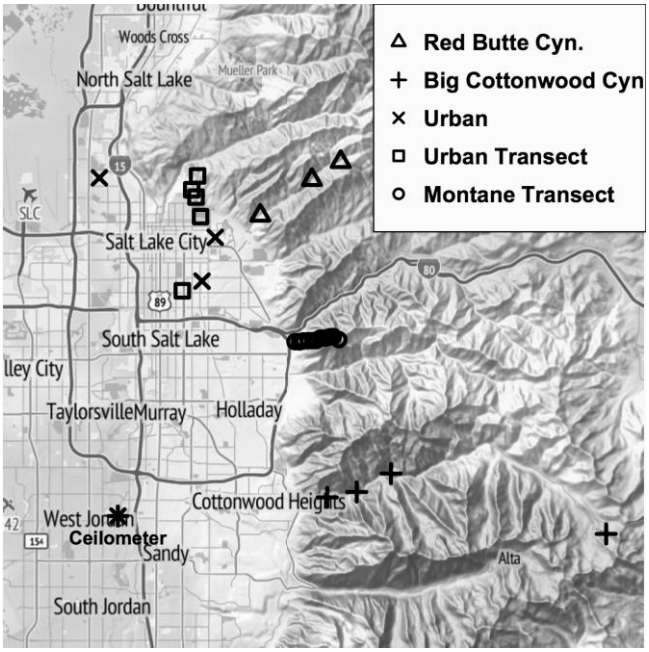

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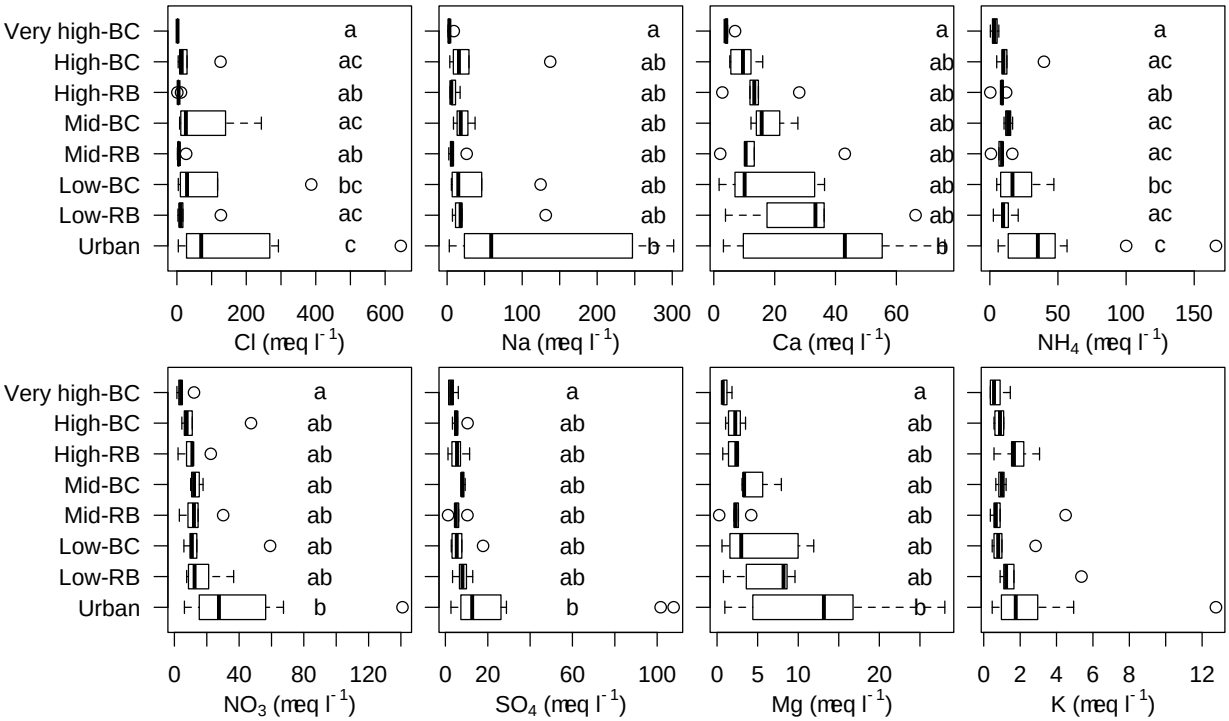
Figure 3:



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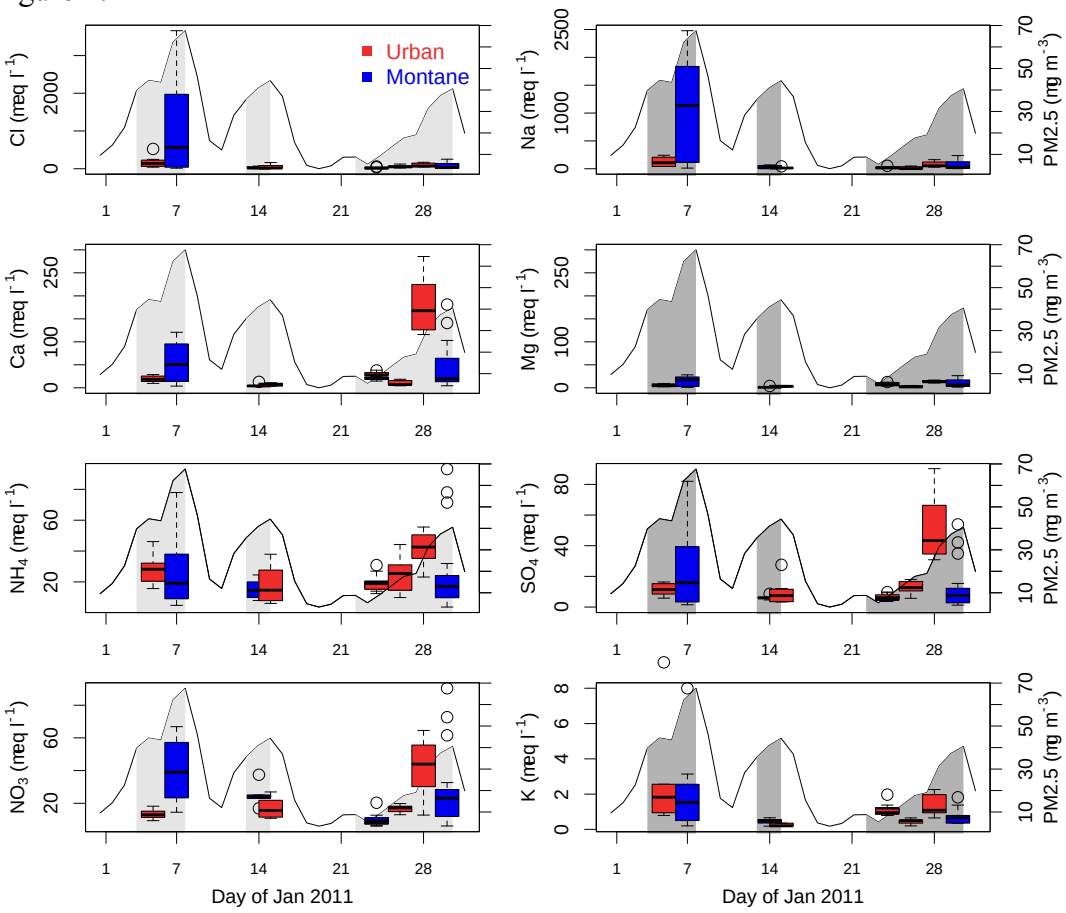

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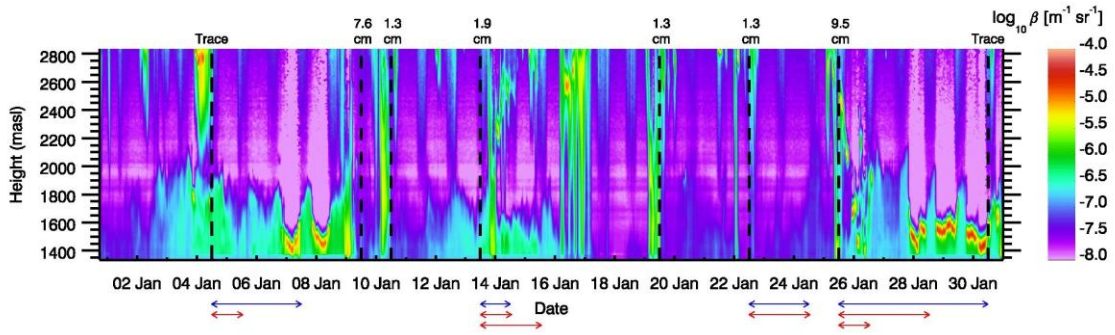

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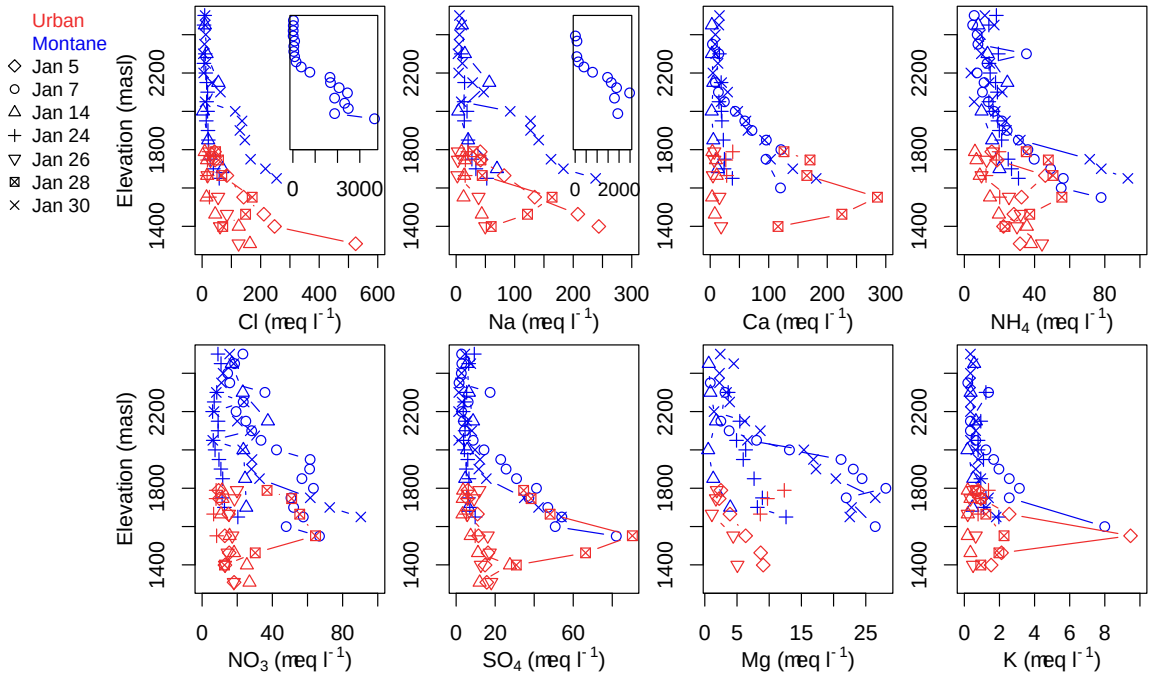

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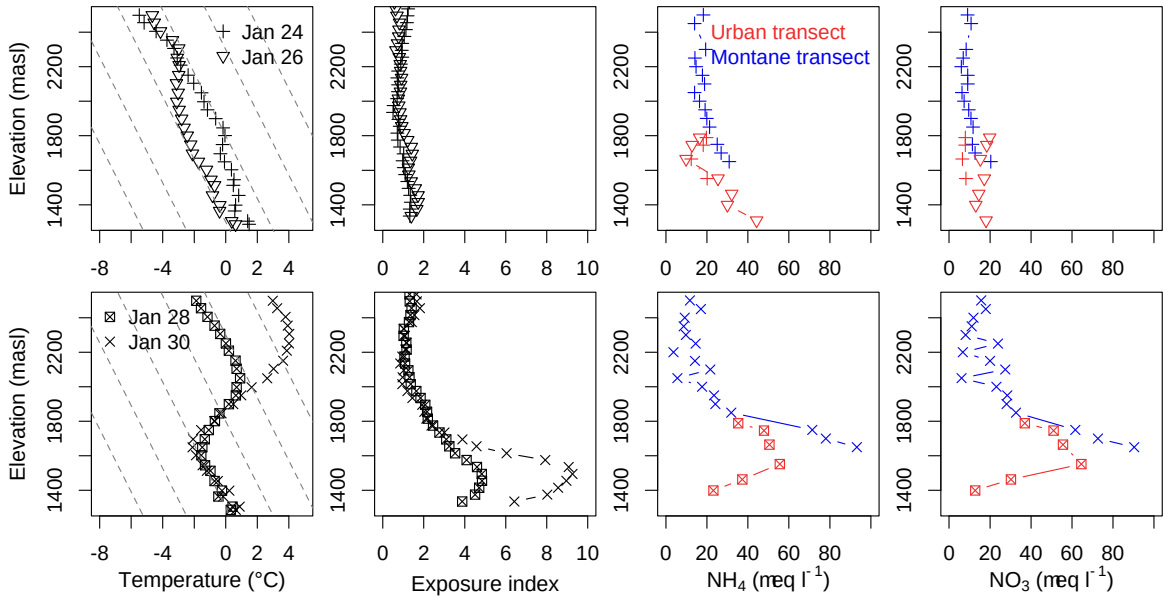
725 Figure 7:



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Appendix A

Figure A1:

Ion loading (the product of ion concentrations and snow water equivalent) in bulk snow samples by site arranged by elevation on the ordinate. Each datum represents an individual snow precipitation event. Boxes represent medians and the interquartile range; data > 1.5 times the range from the box to the whiskers are denoted as outliers (circles). Urban denotes samples from three urban locations, and low, mid, high, and very-high prefixes represent site elevation in Red Butte (RB) and Big Cottonwood (BC) canyons, respectively.

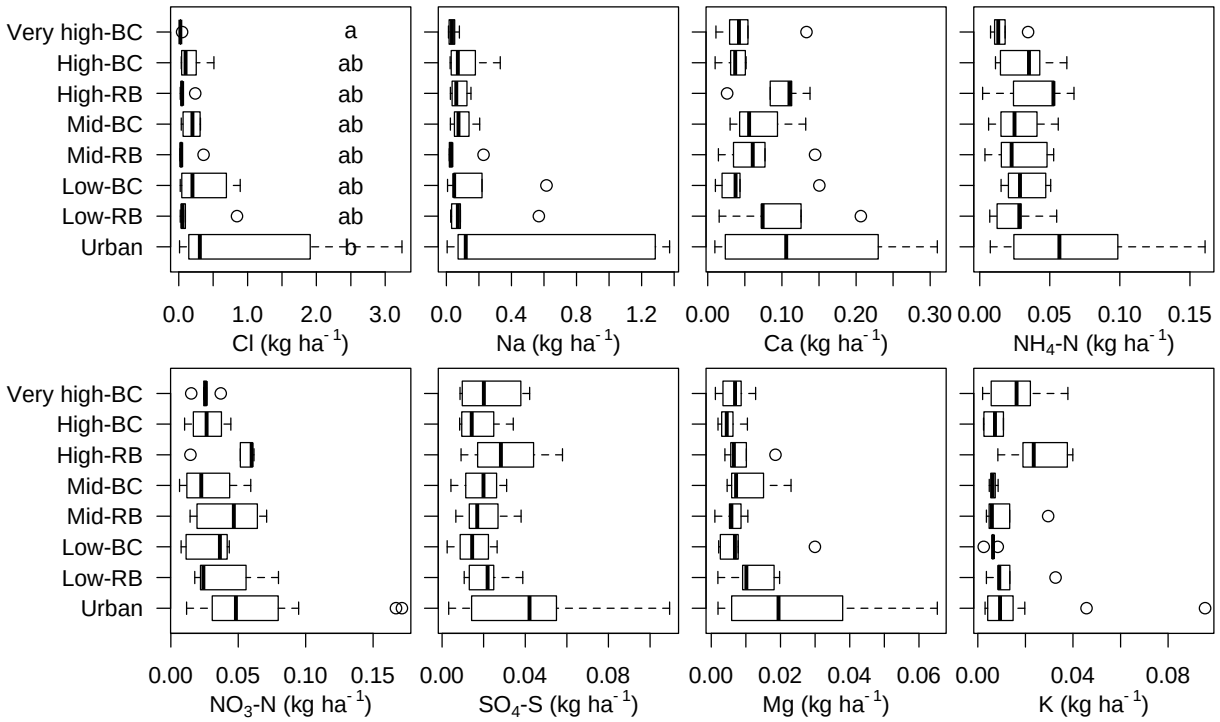
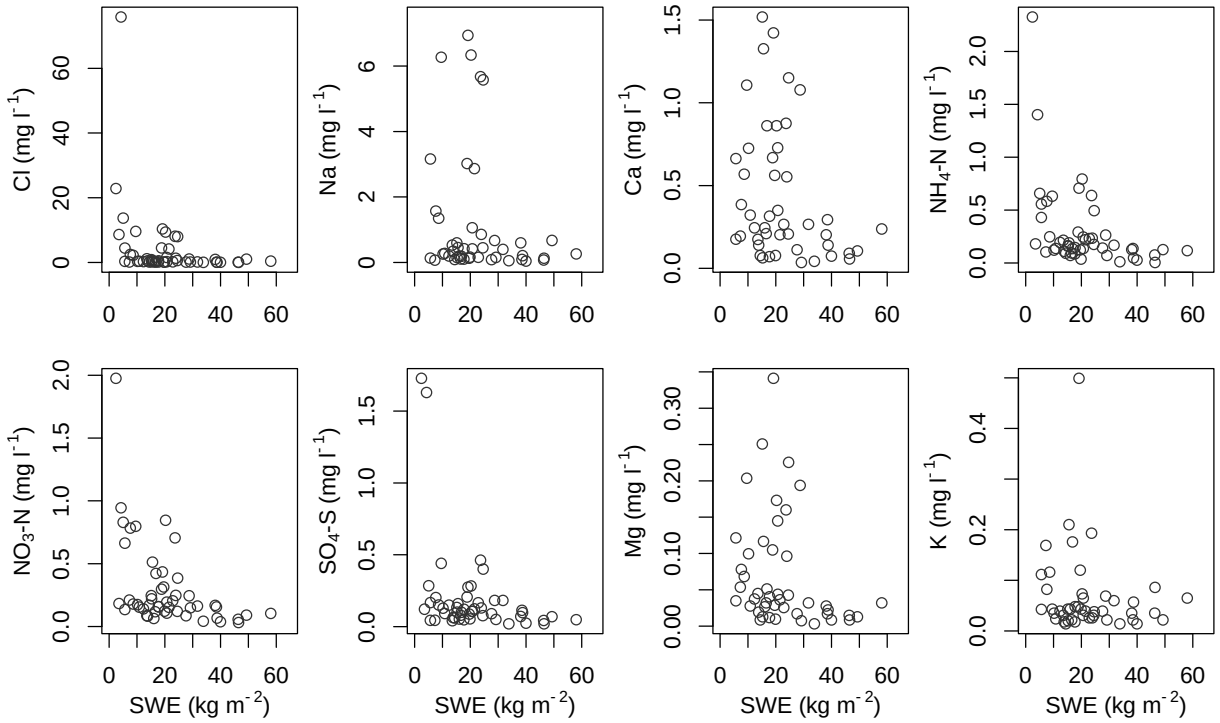


Figure A2:

Relationships between ion concentrations and snow water equivalent (SWE) in the bulk snow samples.



757 Figure A3: Patterns of aerosol exposure (see Methods) plotted by elevation and time during the
758 development of two cold air pools (top and bottom panels) in January 2011. Different symbols
759 represent different dates as indicated in the legends.

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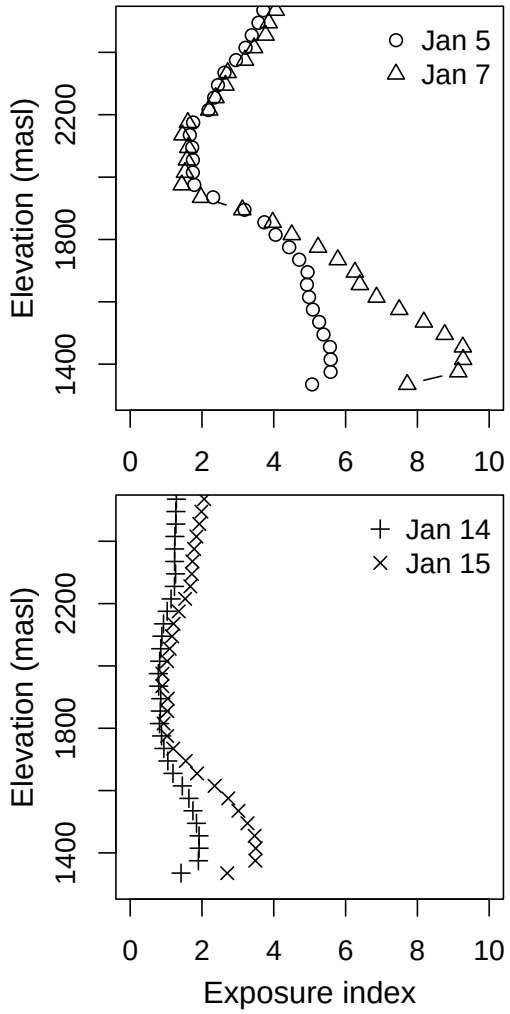

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