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Variations in Geochemistry and Mineralogy of Aeolian Dust Deposition

to Mountains in Utah and Nevada, USA

Dylan Binder Dastrup

A thesis submitted to the faculty of
Brigham Young University
in partial fulfillment of the requirements for the degree of

Master of Science

Gregory T. Carling, Chair
Stephen T. Nelson
Barry Bickmore

Department of Geological Sciences
Brigham Young University

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ABSTRACT

Variations in Geochemistry and Mineralogy of Aeolian Dust Deposition to Mountains in Utah and Nevada, USA

Dylan Binder Dastrup
Department of Geological Sciences, BYU
Master of Science

Major and trace metal loading to mountains in the western US depends on dust sources, intensity of storms and their availability for transport during snowmelt and runoff. Previous work has been conducted on dust production, composition, and its affect on solar radiation and timing of snow melt. This study was conducted to 1) examine temporal and spatial variability in dust chemistry; 2) evaluate form and availability of major and trace elements in dust; and 3) identify potential dust sources affecting mountains in Utah and Nevada. Spring and summertime dust was collected across northern Utah over the course of three years (2013-2015). Additional dust samples were collected from eastern Nevada for comparison. All samples were analyzed for mineralogy. The spring dust samples were also leached with 1 M acetic acid, 0.8 M nitric acid, and aqua regia and analyzed for $^{87}\text{Sr}/^{86}\text{Sr}$ ratios and concentrations of 40+ trace and major elements. Nearly all dust samples were enriched in playa-associated elements (U, Mg, Li, Ca, Sr, As) and anthropogenic elements (Sb, Mn, Zn, Cu, Pb, Se, Cd) relative to average upper continental crust. Leachate results showed that nearly 60% Ca, Sr, and Cd mass is potentially available for transport during snowmelt and that the rare earth elements could be mobilized under lower pH conditions in the soil zone. A major dust event on 17 March 2014 that was sampled across the study area showed spatially variable trace element concentrations and $^{87}\text{Sr}/^{86}\text{Sr}$ ratios, indicating that dust deposited to mountain snowpack originated from multiple upwind desert dust source areas. The NOAA HYSPLIT model was used to calculate back trajectories for this dust event and showed potential dust sources ranged from the Sevier, West and Great Salt Lake deserts in Utah and the Snake River Plain in Idaho. In contrast, multivariate statistical analysis showed that over the course of the study samples had unique geochemical signatures within each sample area. These findings suggest that spatial variability is more important than temporal variability in terms of the chemistry of dust deposition. With increasing populations and land use change in the western US, the short and long term effects of aeolian dust deposition to mountain environments need to continual monitored and constrained.

Keywords: snow, dust, geochemistry, isotope, mineralogy

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Introduction

Aeolian (wind-blown) dust is potentially an important source of nutrients and major/trace elements to mountain ecosystems worldwide (Munroe, 2014; Munroe et al., 2015; Reynolds et al., 2014; Reynolds et al., 2010). In the western US, dust deposition has increased 500% over the past century, resulting in increased deposition of K, Mg, Ca, N, and P to parts of the Rocky Mountains (Neff et al., 2008). The increased dust flux is driven by human-caused disturbance of desert soils, including the expansion of agriculture, livestock grazing, mining operations, and post-fire treatments (Belnap and Gillette, 1998; Mahowald et al., 2010; Miller et al., 2012; Moulin and Chiapello, 2006; Neff et al., 2005; Reynolds et al., 2010). Over the past two decades dust deposition has caused accelerated Ca^{2+} deposition and increased alkalinity of surface waters across the western US (Brahney et al., 2013). Continued groundwater withdrawal in desert areas, such as the proposed “water grab” in Snake Valley Utah-Nevada (Gillespie et al., 2012), has the potential to only exacerbate dust emissions as already dry areas become new dust bowls like Owens Dry Lake and other anthropogenic-disturbed playas (Gill et al., 1999; Reheis et al., 2002; Reheis et al., 2009). Regardless of direct human impacts, dust emissions in the southwest US are expected to continue to increase due to climate change-driven increase in aridity and decrease in perennial plant cover (Field, 2009; Munson et al., 2011; Woodward et al., 2005).

Dust contributes substantial loading of soluble salts, metals, and metalloids to snowpack (Carling et al., 2012; Clow et al., 2002; Ingersoll et al., 2008; Lawrence et al., 2010; Rhoades et al., 2010; Turk et al., 2001). For example, major ion concentrations (Ca^{2+} , Na^+ , Cl^- and SO_4^{2-}) were two to three-fold higher in snow containing dust relative to dust-free snow in the San Juan Mountains in Colorado (Lawrence et al., 2010). Similarly, trace metal/metalloid concentrations in the Wasatch

Mountains in Utah were up to five-fold higher in dust-containing snow relative to dust-free snowpack (Carling et al., 2012). Trace metal concentrations in dust (e.g., As, Cd, Cu, Mo, Pb, and Zn) are typically enriched relative to average upper continental crust, and dust-derived trace metals are more available and mobile relative to other sources (Lawrence and Neff, 2009; Lawrence et al., 2010; Lawrence et al., 2013). Dust contributes both soluble species and particulate matter to snowpack, with potential differences in fate and transport during snowmelt (Bacardit and Camarero, 2010; Bohdalkova et al., 2012; Deuerling et al., 2014; Fortner et al., 2011; Gaspari et al., 2006; Grotti, 2011). Dust can also affect the radiation balance of snowpack, leading to earlier melt (Painter et al., 2007; Painter et al., 2010).

Dust composition is typically characterized using a variety of isotopic, mineralogical, and geochemical methods, which together provide information on dust sources and anthropogenic inputs. The Sr isotopic ratio ($^{87}\text{Sr}/^{86}\text{Sr}$) of dust depends on the geology of the dust emission sources and because the $^{87}\text{Sr}/^{86}\text{Sr}$ ratio undergoes minimal fractionation during transport it is a useful tool for tracking dust sources (Miller, 2013). For example, $^{87}\text{Sr}/^{86}\text{Sr}$ ratios of Sr in carbonate minerals have been used to distinguish between playa dust sources (Marshall, 1994) and as tracers of the fate of dust-derived Sr in mountain watersheds (Clow et al., 1997). Dust mineralogy can be used to identify spatial and temporal differences between samples and possible source areas (Munroe, 2014; Shao et al., 2008). Geochemical analysis through sequential leaching provides an estimate of the partitioning of elements among a variety of geochemical and mineral phases (Carling et al., 2012; Lawrence et al., 2010), which mimics the availability of major and trace elements to the environment. Major and trace element enrichment relative to average upper continental crust (UCC) provide evidence of possible dust sources and

aid in identifying anthropogenic inputs (Lawrence et al., 2010; Schütz and Seibert, 1987; Trzepla-Nabaglo, 2007).

Dust-mediated deposition of trace metals to mountain snowpack and soils needs to be characterized to predict potential impacts to alpine ecosystems. Mountain snowmelt is a critical resource for sustaining ecosystems and providing regional water supplies across the western US. Dust deposition potentially adversely affects mountain snowmelt which compromises water quality and quantity throughout the western US. For example, in northern Utah, USA, seasonal mountain snowpack is the primary water source for over 80% of the state's population. A greater understanding of dust sources and the impacts of trace and major elements on mountain ecosystems would motivate better land management practices for reducing dust emissions from source areas. In this study, we investigated dust deposition across four areas in northern Utah and one area in eastern Nevada. The goal of this research was to: 1) examine temporal and spatial variability in dust chemistry; 2) evaluate form and availability of major and trace elements in dust; and 3) identify potential dust sources affecting mountains in Utah and Nevada.

Methods

Sample collection

Dust samples were collected from snowpack in the Uinta Mountains (hereafter Uintas), the Wasatch Mountains (hereafter Wasatch), and the Snake Range at Great Basin National Park (hereafter GBNP) (Figure 1). The Uintas are located in northeastern Utah and run parallel to the Utah-Wyoming border and are the furthest of the sampling locations from desert dust sources. The Wasatch are located in northern Utah and run north-south from the Utah-Idaho border to central Utah on the eastern edge of the Great Basin region. Samples from the Wasatch were

collected in the north (near the Utah-Idaho border), central (near Salt Lake City), and south sections (near Provo City). GBNP is located on the Utah-Nevada border in the central part of the Great Basin region.

Complex meteorology in the Great Basin region produces dust from a variety of sources. Dust sources include lakebed sediments and salt pans (hereafter “playas”) that are remnants of Pleistocene Lake Bonneville (Currey, 1990). Dust storm events in Utah occur primarily during early spring coinciding with peak snowpack accumulation in the mountains (Carling et al., 2012; Hahnenberger and Nicoll, 2012). Dust storms exhibit a diurnal cycle and are most common in the late afternoon and evening hours with the greatest dust fluxes being accompanied by strong southerly winds and to a lesser extent, winds from the northwest (Hahnenberger and Nicoll, 2012; Steenburgh et al., 2012).

We sampled a total of five dust layers from snowpack during spring 2014 and spring 2015. The dust events occurred on 6 February 2014, 17 March 2014, 22 April 2014, 5 April 2015 and 14 April 2015 (hereafter identified as dust events 1 through 5, respectively) (Table 1). Dust layers were sampled opportunistically from snowpack because snow offers a clean surface for dust to land on and also limits entrainment of local dust. Not all dust events affected all of the sampling areas and thus not all sites were sampled after each dust event. Dust event 2 (17 March 2014) was an exceptional event that was sampled at all sites, thereby providing an opportunity to compare the composition of dust from a single event across a large area. All dust events were sampled at the central Wasatch site allowing for a comparison of temporal variability in dust composition at a single sample location. Additionally, dust events 3 (22 April 2014) and 5 (14 April 2015) were sampled at the western Uintas site. Because dust was deposited to snowpack and sampled prior to the onset of melt, the samples represent unmixed single dust event layers

that can be correlated with specific regional wind events.

To collect the maximum amount of dust from each layer, snow dust was collected by digging a pit to the ground surface and scraping the mouth of a 2.5 L FLPE acid leached bottle across an exposed dust layer either on the snow surface or along the snowpit wall. Samples were collected while wearing powder-free vinyl gloves and Tyvek lab coats. ‘Clean hands, dirty hands’ methods set by the EPA were observed while sample collection took place (USEPA, 1996). Bottles were transported and stored frozen until processing in the laboratory.

To evaluate seasonal variability in dust mineralogy, dust buckets were used to sample summertime dust deposition in the absence of snowpack. The dust bucket design was modified from collectors described in Reheis et al. (1995). Dust buckets were constructed using plastic HDPE three gallon buckets, lined with a 4 mm clear polyethylene bag. A plastic screen with glass marbles was placed about 10 cm from the rim of the bucket to prevent wind from removing dust from the collector. Dust buckets were deployed in the western Uintas, central Wasatch, southern Wasatch and at GBNP (Figure 2) during summer 2013 and 2014 (Table 2). Because the dust buckets were deployed over a period of several months (or longer), the accumulated dust could not be correlated to specific wind events.

Dust buckets were deployed from September to October 2013 and July to October 2014 in the southern Wasatch and western Uintas (Table 2). At GBNP dust buckets were deployed from September 2013 to July 2014 and July 2014 through October 2014 (Table 2). Dust buckets were placed in open areas, on hilltops and in meadows away from trees and other potential local dust sources. The first set of dust buckets most likely captured snowmelt and dust that may have fallen during the winter and spring and mineralogy from those samples will represent any

mixture of all deposited dust. Dust buckets were a modified design of collectors used by Reheis et al. (1995). Dust buckets were constructed using plastic HDPE three gallon buckets, lined with a 4 mm clear polyethylene bag. A plastic screen with glass marbles was placed about ten cm from the rim of the bucket to prevent wind from removing dust from the collector. Upon retrieval any large debris was removed from the marbles using vinyl powder free gloves and marbles were rinsed with one liter of ultra pure water to wash the dust down into the bucket. The 4mm plastic bag was sealed with tape and the buckets were sealed with GAMA seal lids for transportation.

Sample preparation

Prior to chemical and mineralogical analysis snow dust samples were isolated from the melted snow. Bottles for snow dust samples were rinsed with ultra pure water and left to dry in a laminar hood while samples thawed, which usually took about 24 hours. Each bottle was agitated by hand for ~1 minute and the sample was poured into 50 mL centrifuge tubes. The tubes were centrifuged at 3000 RPM for 5 minutes and the supernatant was poured off. This step was repeated until the dust from an entire sample was composited into one 50 mL centrifuge tube with the dust on the bottom. The remaining sample was air-dried in a laminar flow hood. It is possible that pouring off the supernatant likely resulted in a loss of soluble ions (Na^+ , K^+ , Cl^-) in the dust samples but measuring the soluble salts was not considered necessary for the purposes of this study.

Dust samples from dust buckets were a mixture of rainfall and ultrapure rinse water. Water was decanted from the dust buckets until around two liters of sample remained which was then transferred to 2.5L FLPE bottles and allowed to settle for 24 hours. The samples were again

decanted until ~200 mL of sample remained. The remaining ~200 mL was composited into a 50 mL centrifuge tube by spinning the sample at 3000 RPM for 5 minutes and decanting the supernatant. The remaining sample was air-dried in a laminar flow hood. To remove any organic material that may have grown or fallen into the sample buckets during deployment, 30 mL of 30% H₂O₂ was added to each sample and heated at 60° C for one week. Samples were then centrifuged at 3000 RPM for 5 minutes and the supernatant was poured off. Samples were rinsed by mixing 30 mL of ultra pure water with the sample, centrifuging at 3000 RPM for 5 minutes then pouring off the supernatant. Samples were dried in a laminar flow hood by placing the centrifuge tubes into quartz sand heated to 60° C for 72 hrs.

Sequential leaching

To assess the distribution of elements among unique chemical fractions, snow dust samples were subjected to a multistep leaching process prior to geochemical analyses. The goal of this leaching process was to isolate elements from the exchangeable/carbonate (hereafter exchangeable) fraction (i.e., elements that are most likely mobile during snowmelt and in slightly acidic conditions in the soil) (Litaor et al., 2004), organic matter, feldspar and clay fractions (hereafter organic), and residual element fractions (Carling et al., 2012; Lawrence et al., 2010). For the first leaching step, 3 mL of 1 M acetic acid was added to ~20 mg of dust in a clean 15 mL centrifuge tube, agitated on a vortex and leached for 24 hours. The sample was centrifuged at 3000 RPM for 10 minutes and 2 mL of leachate was combined with 3 mL of 0.8 M HNO₃ in an acid-washed 15 mL polypropylene sample tube for element analysis. The remaining supernatant was removed and the dust was rinsed by mixing it with ultra pure water, centrifuging at 3000 RPM for 10 minutes and decanting the rinse water. For the second leaching step, 3 mL of 0.8 M HNO₃ was added to the remaining dust, mixed on the vortex, and allowed to leach for 24 hours. The sample

was centrifuged at 3000 RPM for 10 minutes and 2 mL of leachate was combined with 3 mL of 0.8 M HNO₃ for analysis. For select samples, 5 mL of aqua regia was added to 20 mg of non-leached dust in a clean 15 mL centrifuge tube, agitated on a vortex and leached for 24 hours. The sample was centrifuged at 3000 RPM for 10 minutes and 1 mL of leachate was diluted to 10 mL with 0.8 M HNO₃ for analysis. Because a non-leached dust sample was used for the aqua regia leach, the residual fraction was calculated by subtracting the measured concentrations of the exchangeable and organic fractions from aqua regia concentrations.

Laboratory analysis

Major and trace element concentrations were measured from the acetic acid, nitric acid, and aqua regia leachate fractions. Concentrations were measured for the following 40 elements: Ag, Al, As, B, Ba, Be, Ca, Cd, Ce, Co, Cr, Cs, Cu, Dy, Eu, Fe, Gd, Ho, K, La, Li, Lu, Mg, Mn, Mo, Na, Nd, Ni, Pb, Rb, Sb, Se, Sm, Sr, Tb, Ti, Tl, U, V, Y, and Zn. Samples were analyzed using an Agilent 7500ce quadrupole inductively coupled mass spectrometer (ICP-MS) with a collision cell, a double-pass spray chamber with perfluoroalkoxy (PFA) nebulizer (0.1 mL/min), a quartz torch, and platinum cones. The detection limit (DL) was determined as three times the standard deviation of all blanks analyzed throughout each run. A USGS standard reference sample (T-205) and NIST standard reference material (SRM 1643e) were analyzed multiple times in each run together with the samples as a continuing calibration verification. The long-term reproducibility for T-205 and SRM 1643e show that our results are accurate within 10% for most elements.

Snow dust samples were analyzed for ⁸⁷Sr/⁸⁶Sr ratios using a Thermo Scientific Neptune multicollector ICP-MS by the same methods as Carling et al. (2015). The samples were purified

inline using a Sr-FAST ion chromatographic column packed with a crown ether resin (Mackey, 2011). During the analyses reported herein, we determined the $^{87}\text{Sr}/^{86}\text{Sr}$ ratio of the standard reference material SRM987 ($n = 20$) to be 0.71030 ± 00001 (2σ standard error (SE)). Analytical precision (2σ SE) of all samples ranged from ± 00001 - 00003 . The $^{87}\text{Sr}/^{86}\text{Sr}$ ratios were corrected for mass bias using an exponential law, normalizing to $^{87}\text{Sr}/^{86}\text{Sr} = 0.1194$ (Steiger and Jäger, 1977). Isobaric interferences on the $^{87}\text{Sr}/^{86}\text{Sr}$ ratios, such as from ^{87}Rb and ^{86}Kr , were corrected by simultaneous monitoring of ^{87}Rb and ^{86}Kr using the corresponding invariant ratios of $^{87}\text{Rb}/^{86}\text{Rb} = 0.385706$ and $^{86}\text{Kr}/^{83}\text{Kr} = 1.502522$ (Steiger and Jäger, 1977).

Dust mineralogy was analyzed by x-ray diffraction (XRD) using a PANalytical X'Pert Pro MPD. In preparation for XRD analysis a clean stainless steel spatula was used to break up any clumps of sample that may have formed in the centrifuge tube. The powder was then transferred to a zero background holder and compressed as flat as possible. Samples were analyzed by diffractometer scanning from 2° to 65° with a step of 0.008° and the resulting XRD patterns were analyzed using the Rock Jock V.11 software (Eberl, 2003). Rock Jock sample preparation procedures (Eberl, 2003) were not strictly followed, which may decrease accuracy and precision, but the modified method was necessary because of the small sample volume and the use of zero background holders. Because of a lack of sample volume not all samples were analyzed for mineralogy.

Data analysis and modeling

Non-metric multidimensional scaling (NMS) was used to explore and visualize the spatial and temporal variability among snow dust samples. NMS is a data ordination technique that can be used to explain relationships of elemental composition by ordering objects (samples) along axes

according to their similarities (McCune, 2002). Although there are various methods of ordination, NMS proves to be robust in that it avoids the assumption of linear relationships and normal distributions among variables (Peck, 2010). NMS was performed using the PC-ORD (McCune, 1999) software package, categorizing samples by sample location. The data were prepared for NMS through log generalization using the equation $b = \log(x + x_{\min}) - \log(x_{\min})$, where x_{\min} is the minimum positive value for the element (column). Log generalization rescales the values in such a way that very large or very small values are brought much closer to the mean, which reduces the range in value and allows for the analysis of samples with a broad range of values.

Through the NMS analysis, a distance matrix was constructed for each value from the original dataset using Euclidean distances measures. The distance between pairs of values were ranked and became the target rank order. Random ordination scores were then assigned to each sample unit and the distance between each value was calculated again. The distance between pairs of the reduced values were ranked and ordered. The relationship between the target rank order and the reduced rank order were compared and should be monotonically nondecreasing. Ordination scores were adjusted and the process repeated until monotonicity is achieved. The amount of adjustment needed is known as “stress” where smaller amount of adjustment produces lower stress levels. A final stress < 10 is acceptable (Peck, 2010). The analysis was performed 250 times with the lowest stress level being chosen as the final solution. Ordination values from the final solution were plotted in ordination space. These values had no intrinsic meaning and only reflect a model of variation that was present in the original data. Sample locations were grouped using polygons called “convex hulls”. Objects that were close in ordination space are generally more similar than objects distant in ordination space (McCune and Mefford, 1999). Post-hoc

analysis of the coefficients of determination for the correlations between ordination distances and distances in the original n-dimensional space were used to estimate the amount of variability in the data explained by each of the ordination axes.

NMS was performed for each leach fraction (acetic acid, nitric acid, and aqua regia) using Al, As, Ba, Be, Ca, Cd, Ce, Co, Cr, Cu, Dy, Eu, Fe, Gd, Ho, K, La, Li, Lu, Mg, Mn, Na, Nd, Ni, Pb, Rb, Sb, Sm, Sr, Tb, Ti, Tl, U, V, Y, and Zn. Silver, B, Mo and Se were removed from analysis because they were below detection limit in the majority of samples. Cesium was removed from the final analysis because of the potential contamination of one of the samples from the central Wasatch during 2015. For other elements, values at or below detection limit were set to ½ the detection limit for NMS analysis.

To investigate possible dust sources using meteorological data, the National Oceanic and Atmospheric Administration (NOAA) Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT) model was used to calculate 24 hr back trajectories (Draxler, 2004). HYSPLIT was run using NARR 32 km model meteorological data. READY (Real-time Environmental Applications and Display sYstem) provided by the NOAA Air Resources Laboratory (ARL) was used to run each back trajectory (NOAA, 2011). Trajectories were run for 12 h at a height of 500 m above ground level (AGL).

Results

Mineralogy of snowpack and dust bucket samples

Mineralogy of snowpack dust and dust bucket samples show spatial and temporal variability across the sampling area (Figure 2). All of the snow dust samples contained quartz (ranging from 18 to 48% abundance) and nearly all samples contained plagioclase (up to 30%) and illite clays

(up to 20%). The most important differences in snowpack dust mineralogy were the relative abundance of K-feldspar and calcite across the sample sites. The northern Wasatch, western Uinta, and GBNP samples contained relatively high abundance of K-feldspar (20 – 30%) whereas the central and southern Wasatch samples contained no measureable K-feldspar. Calcite was found in a majority of samples from the central and southern Wasatch and Uinta samples (up to 5% abundance) but not in the northern Wasatch and GBNP samples. Thus, K-feldspar and calcite may serve as diagnostic minerals for differentiating dust across the study area. Taken together, three distinctive groups were found in the mineralogy of snow dust samples for the dust event 2 on 17 March 2014: 1) the northern Wasatch and GBNP samples (contain K-feldspar but not calcite); 2) the central and southern Wasatch (contain calcite but not K-feldspar); and 3) western Uintas (contain K-feldspar and calcite). Mineralogy at the central Wasatch and Uinta sites were similar over time. It is not clear whether the mineralogy at the northern Wasatch and GBNP samples are consistent over time since samples were only collected at these locations for dust event 2, but this is an area for future research.

All dust bucket samples contained quartz (ranging from 23 to 55% abundance) and nearly all samples contained smectite or illite clays (up to 44%). The noticeable difference between the samples is the presence or absence of K-feldspar or plagioclase. The western Uinta samples from 2013 had an abundance of plagioclase (up to 28%) and illite clays (up to 25%), but K-feldspar was only found in one sample. In contrast, the 2014 samples from the western Uintas contained K-feldspar and smectite but no measurable amount of plagioclase. The southern Wasatch samples from 2013 contained K-feldspar (up to 29%) but no measureable amount of plagioclase. The 2013 samples from GBNP contained plagioclase (up to 19%) and illite (up to 27 %) whereas the 2014 samples had K-feldspar (up to 41%) and calcite (up to 14%) with no measurable

amount of plagioclase. The dust bucket samples can be classified into four groups: 1) 2013 samples that are K-feldspar dominated, southern Wasatch 1 and 2; 2) 2013 samples that are plagioclase dominated, western Uintas 1,2 and 3, GBNP 1A and 2A; 3) 2014 samples that are K-feldspar dominated, western Uintas 5 and 6, GBNP 1B; and 4) 2014 samples with no measureable K-feldspar or plagioclase, which ones?.

The relative abundance of minerals in the dust bucket samples was dominated by one or two minerals, whereas the snow dust samples were more evenly dispersed. Differences in mineralogy between the snow dust samples and dust bucket samples reflect seasonal differences in dust deposition between spring and summer months, respectively. Snow dust had a greater diversity of minerals, but less variability between samples than the dust bucket samples. Calcite was more abundant in the snow dust samples (central Wasatch, southern Wasatch and western Uintas) than the dust bucket samples (GBNP and southern Wasatch). Overall, the majority of the dust bucket samples contained more quartz than the snow dust samples. Western Uinta snow dust and dust bucket samples from 2014 both had quartz, K-feldspar, smectite and illite, but the snow dust samples also had calcite and plagioclase.

Sequential leach percentages

Sequential leaching of dust samples shows variability in the partitioning of elements between the exchangeable, organic and residual phases (Figure 3). The central and northern Wasatch samples represent end members for the range of values seen during the study. For example, greater than 90% of the Ca and 70% of Na from the central Wasatch were made available from the acetic acid leach where only 70% of the Ca and 40% of Na were made available from the northern Wasatch samples. For all samples nearly 60% of Sr and Cd were removed in the acetic acid leach,

compared with only 20% of Mg, Na, Mn, Zn and Be. A large fraction (~60%) of Cr and all REE were removed with the nitric acid leach. It is possible these elements were associated with large organic particles in the dust samples (Lawrence et al., 2010). For the northern Wasatch and central Wasatch, other elements including Al, Fe, Ti, B, Rb, V, Ni, As, Mo, Cs Sb, Tl and Ag were leached with aqua regia (Figure 3). Overall, the central Wasatch samples had a higher availability of all elements from the acetic and nitric acid leached relative to the northern Wasatch samples.

$^{87}\text{Sr}/^{86}\text{Sr}$ ratios

Considering the samples collected at all sites for dust event 2 on 17 March 2014 (Figure 4A), the northern Wasatch samples had the lowest $^{87}\text{Sr}/^{86}\text{Sr}$ values (ranging from 0.709 in the nitric acid leach to 0.710 in the acetic acid leach) and central Wasatch samples had the highest $^{87}\text{Sr}/^{86}\text{Sr}$ values (0.711 – 0.712). Intermediate values were found in samples from the southern Wasatch, western Uintas, and GBNP with $^{87}\text{Sr}/^{86}\text{Sr}$ values ranging from 0.710 to 0.711. Overall, $^{87}\text{Sr}/^{86}\text{Sr}$ ratios were less radiogenic in the acetic leach fraction relative to the nitric leach fraction. The northern Wasatch samples had the greatest difference between the $^{87}\text{Sr}/^{86}\text{Sr}$ ratios of the acetic and nitric leaches (0.0010 ± 0.00005 , $n = 3$) relative to samples from the other sites samples (0.0006 ± 0.0003 $n = 12$). Samples from the southern Wasatch and GBNP showed similar $^{87}\text{Sr}/^{86}\text{Sr}$ ratios in the nitric and acetic acid leach fractions, reflecting potential differences in mineralogy or anthropogenic inputs relative to samples from the other sites. Through analysis of $^{87}\text{Sr}/^{86}\text{Sr}$ ratios for dust event 2, samples can be separated into two distinct groups, 1) the northern Wasatch and 2) central Wasatch, southern Wasatch, western Uintas and GBNP.

Samples from dust events 1 – 5 collected at the central Wasatch site (Figure 4B) show $^{87}\text{Sr}/^{86}\text{Sr}$ ratios varied over the course of the study. Samples from dust events 1, 2 and 5 are less depleted in both leach fractions than samples from dust events 3 and 4. Although each value from the acetic leach was different from one another, the values for the nitric fractions were bimodal. The central Wasatch samples can be separated into three groups based on $^{87}\text{Sr}/^{86}\text{Sr}$ ratios: 1) dust events 1, 2 and 5, which have similar isotopic ratios and an average difference between the two leaches of 0.00079 ± 0.0002 $n = 3$; 2) dust event 3, which was less radiogenic relative to 1, 2, and 5 and both leach fractions had nearly identical ratios; and 3) dust event 4, which was also less radiogenic relative to 1, 2, and 5 but showed differences between acetic acid and nitric acid leach steps.

Enrichment factors

To evaluate the relative contribution of major and trace elements from natural versus anthropogenic sources, crustal enrichment factors (EF_{UCC}) were calculated by comparing elemental values from the snow dust samples to those of the upper continental crust (UCC) (McTainsh and Strong, 2007; Piper, 2013; Wedepohl, 1995). This ratio was calculated by the equation $\text{EF}_{\text{UCC}} = ([\text{X}]_{\text{s}}/[\text{Al}]_{\text{s}})/([\text{X}]_{\text{UCC}}/[\text{Al}]_{\text{UCC}})$, where X is the element of interest and the subscript s denotes concentration in the sample. An enrichment factor of 0.1 – 10 represents crustal composition whereas values >10 indicate that the sample is enriched relative to average continental crust. Enrichment factors with a range of 10 – 500 indicate moderate enrichment and values greater than 500 suggest anthropogenic inputs.

Enrichment factors calculated for snow dust deposited during dust event 2 (17 March 2014) show that all samples were moderate to highly enriched in playa-associated (U, Li, Mg, Ca, Sr,

B) and anthropogenic elements (Sb, Mn, Zn, Cu, Pb, As, Se, Cd) in the acetic acid leach (Figure 5). All samples were greatly enriched in Cd with EF_{UCC} values over 500. The exception was B, which was only moderately enriched in the central Wasatch, southern Wasatch and western Uintas samples. The central Wasatch, southern Wasatch and western Uintas were more greatly enriched in playa and anthropogenic elements than the northern Wasatch samples.

Elements leached from nitric acid are associated with organic material in the dust. Silver, Li (except for the northern Wasatch) Zn, Cu, Pb, As, Se and Cd were moderately enriched in all samples from dust event 2. All elements from the nitric acid leach were less enriched than they were in the acetic acid leach with the exception of Ag. The aqua regia leach revealed that Ag, B, Sb, Pb, As, Se and Cd were moderately enriched in nearly every sample from dust event 2.

Snow dust samples collected from five dust events at the central Wasatch site were only moderately enriched in the exchangeable fraction for some playa and anthropogenic elements (Li, Mg, Ca, Mn, Zn, As and Se) (Figure 6). The exception was Cd, which was highly enriched in all dust events. The two samples from 2015 (4-CW and 5-CW) showed greater enrichment of playa and anthropogenic elements in the exchangeable and organic fractions than the samples from 2014. The aqua regia leach was not performed on all snow dust samples collected at the central Wasatch site, which make it impossible to comprehensively compare EF_{UCC} between the exchangeable, organic and residual fractions.

Overall, REE from all dust events coincided with the average concentration of REE from the upper continental crust (Figure 7). EF_{UCC} for REE were greater in the organic fraction than the exchangeable or residual fractions. The 2015 samples were more enriched than the 2014 samples in the exchangeable fraction but less enriched in the organic fraction. There was a slight Eu anomaly in the organic and residual fractions from all sample locations.

NMS

The NMS ordination yielded a two dimensional model (final stress = 5.71) where axis 1 described 80% of the variability in the dataset and axis 2 described 15 % (Figure 8). Samples collected in the same sample location over the course of this study all plot near one another but sample locations plot in different regions of the ordination space (Figure 8). The northern Wasatch samples are defined by Tl and Fe on the positive end of axis 1. The central Wasatch samples are defined by elements associated with carbonate minerals (Ca, Sr, Mg) and heavy metals (U, Cu, Pb) on the left side of axis 1. The western Uintas samples plot in the middle of axis 1 with intermediate concentrations of all elements. Axis 2 shows relationships within the sites based on differences in Sr, Ca, Cu, Fe and Tl.

HYSPLIT back trajectories

Frontal passages produce dust events across the Great Basin region and are often associated with colder temperatures and precipitation (Hahnenberger and Nicoll, 2012). For dust event 2 (17 March 2014) early morning (10:00 UTC) pre-frontal winds (~ 6 m/s) came from the south-southwest. By 14:00 UTC they had increased in intensity (~ 8-9 m/s) and shifted to the west-northwest and by 18:00 UTC they peaked in intensity (~17 m/s) and shifted to the north-northwest as the storm passed. Because there was such little precipitation, dust sources remained dry and were able to produce dust from both pre and post-frontal winds. HYSPLIT back trajectories and local weather patterns for dust event 2 (17 March 2014) suggest that this storm was unique because the prefrontal winds occurred earlier in the day and there was very little precipitation from the storm (Figure 9).

Discussion

Elemental enrichment factors

Elemental concentrations of most major and trace elements in snow dust samples were similar to the average concentrations of the upper continental crust. The notable depletion of Na in all leach fractions may be explained by 1) weathering processes of the soil before dust entrainment (Lawrence and Neff, 2009; Schütz and Rahn, 1982) and 2) the method of isolating the dust by decanting the melted snow from the sample collection bottles prior to analysis. Interestingly Cd is greatly enriched in all samples in the exchangeable fraction. Cadmium is most often associated with phosphate fertilizers in agricultural soils (Amalhay and Roberts, 2014), combustion of coal (Bettinelli et al., 1988) and smelting of Cu (Lukaszewski et al., 1993). Boron is greatly enriched in southern Wasatch and central Wasatch samples but shows no enrichment in northern Wasatch and GBNP samples. Boron is associated with playa minerals and occurs naturally in Great Salt Lake (Tripp, 2009). It is possible elements that were made available in the nitric acid leach and moderately enriched with respect to the upper continental crust were leached from organics already present in the atmosphere through anthropogenic inputs near populated areas and scavenged as the storm passed through. The differences in depletion and enrichment between sample locations suggest different dust sources affect each site.

One of the most striking differences in the central Wasatch samples was the difference in EF_{UCC} of the playa and anthropogenic elements between the 2014 and 2015 dust events (Fig. 9).

Enrichment factors of U, Li, Mg, Ca, Sr, B, Cu, Pb, As and Se were up to five times greater in the 2015 snow dust samples relative to the 2014 samples. The differences in EF_{UCC} between the two years may be attributed to several factors; 1) less precipitation in dust source areas prior to dust events, allowing more dust to be available for transport (Marticorena et al., 1997); 2) greater

wind speeds during the 2015 dust events or; 3) increased emission rates of anthropogenic elements in the urbanized areas upwind of the sample site.

A small negative Eu anomaly was present in the organic and residual fractions of all samples, which indicate the dust is at least partially derived (ultimately) from sources that underwent feldspar fractionation. Particularly in plagioclase, substantial Eu^{2+} may substitute for Ca^{2+} in place of Sr^{2+} . Liquids that have formed where plagioclase is a stable residual phase, or from which plagioclase is crystallized and lost, will tend to be significantly depleted in Eu (McLennen, 1989). There is a slight negative Ce anomaly in all leaches, which may be caused by oxidizing conditions when the sediments were deposited (McLennen, 1989).

Form and availability of major and trace elements in dust.

The most available fraction of dust is represented in the acetic acid leach, which mimics slightly acidic conditions in the soil (~ pH 3.8 – 4.3) (Litaor et al., 2004) and snowmelt. In all samples Ca and Sr were leached preferentially from the exchangeable fraction. These elements are commonly associated with carbonate minerals (Lawrence et al., 2010). The mineralogy for these samples showed there was no measureable calcite in the northern Wasatch, which suggests whatever Ca was present may be associated with playa material. Cadmium was also abundant in the exchangeable fraction, although its concentrations poorly correlated with concentrations of Ca or Sr, which suggests it is not associated with carbonate minerals in the dust. Similar trends in Cd availability were demonstrated by Lawrence et al. (2010) where it was interpreted as being leached from organic matter in the dust. Although C, N and P were not measured in this study, the large percentages trace element availability from the nitric acid leach suggests they are associated with organic matter in the dust (Lawrence et al., 2010). Trace metal concentrations in

dust are often strongly correlated with organic matter content (Lawrence et al., 2010; Reynolds et al., 2014), which may control the transport of trace metals during snowmelt due to complexation with organic ligands associated with dissolved organic matter (DOM) (Davis, 1984). Organic matter may be partially derived from urban and industrial sources upwind as well as anthropogenic sources such as automotive emissions and industrial activities (Reynolds et al., 2014).

With the possibility that elements from the exchangeable and organic fractions are mobilized during snowmelt, the elements associated with the residual fraction (Al, Fe, K, Tl, B, Rb, V, Ni, As, Mo, Cs, Sb, Tl, Ag) are left behind to aid in soil genesis. Aeolian dust has been shown to aid in alpine soil genesis (Lawrence et al., 2013; Munroe et al., 2015).

Samples from different sample locations responded differently to the acid leaches. The central Wasatch has a greater percentage of Ca, Sr, Cd, Se and REE being made available for transport via snowmelt than the northern Wasatch.

Potential dust sources

Analysis of mineralogy, $^{87}\text{Sr}/^{86}\text{Sr}$ isotopes, trace element chemistry and the HYSPLIT model results suggest that northern Utah receives dust flux from several different sources across the eastern Great Basin. Nearly all samples had some amount of quartz, k-feldspar, plagioclase, illite, smectite, and occasionally calcite, which is consistent with aeolian dust mineralogy measured in the southern Colorado, North Pacific Ocean and west-central Africa (Lawrence et al., 2010; Leinen, 1994; Moreno et al., 2006). The size of dust particles can be helpful in distinguishing between local or regional sources with the average size of dust particles becoming smaller the further from the dust source (Lawrence et al., 2010). Samples from local sources tend

to have higher percentages of quartz and feldspars associated with larger amounts of sand and silt sized particles, whereas dust from regional sources will have a higher abundance of phyllosilicates associated with clay sized particles (Lawrence et al., 2010). Relatively high abundances of quartz and feldspars with a smaller component of illite and smectite in the northern Wasatch, southern Wasatch, and GBNP snow dust samples are characteristic of local-regional dust sources. Alternatively, all samples from the western Uintas show a mixture of all the above-mentioned minerals with greater values of smectite, illite and calcite, which is indicative of dust that has traveled greater distances. Nearly all dust bucket samples across the sampling area have high percentages of quartz, k-feldspar and plagioclase, which suggest more localized dust sources during summer and fall months. Because the dust buckets were deployed for such long periods of time each sample represents a mixture of several possible dust events.

Geochemical analysis of snow dust samples shows sites in the study area received dust from different dust emission sources. $^{87}\text{Sr}/^{86}\text{Sr}$ ratios from the northern Wasatch samples (0.709 – 0.710) more closely resemble isotope ratios from the Snake River Plain in Idaho (0.706 – 0.707) (White, 1998), and may represent a mixture of dust from the Snake River Plain and Great Salt Lake Desert. Samples from the other sites are more representative of playa sediments from the Great Salt Lake, Sevier, and West Deserts (0.7103 – 0.7121) (Miller et al., 2014). HYSPLIT back trajectories also show rapidly shifting wind directions during the 17 March 2014, storm, entraining dust from several different dust sources over the course of a few hours. NMS for all dust samples shows sample sites maintained unique geochemical signatures. Variability in geochemistry and mineralogy across the sample locations over the course of this study, suggest differences in dust sources lead to different chemistry at the deposition site, regardless of when the dust event occurs. Additionally sample locations are continuously affected by the same

natural or anthropogenic dust sources. This suggests future changes or development to a single dust emission source may not be seen ubiquitously across the sample areas.

Conclusions

The deposition of aeolian dust is important to the biogeochemical cycling of major and trace elements in montane environments. Understanding how dust deposition may impact mountain ecosystems could be crucial in motivating better land management practices in the southwestern US.

The geochemistry of snow dust was highly variable between sample locations. Mineralogy, geochemistry and $^{87}\text{Sr}/^{86}\text{Sr}$ isotopes indicate that dust collected from each sample location had a unique geochemical signature. Over the course of this study it was determined that there was a greater amount of variation spatially rather than temporally between the samples. This suggests sample locations consistently receive dust from the same dust emission source, but that these sources do not have equal effect on sample locations.

Dusts from each sample area responded differently to sequential leaching and therefore potential environmental conditions. Some dust samples released greater percentages of heavy and trace metals during the leaching process than others. With the increase of dust emissions across the southwestern US due to climate change and increased development upwind, it is inevitable mountains in these regions will continue to receive dust flux in the future. Over time sample areas may receive a disproportional flux of heavy and trace metals from existing or new dust sources, which could create unique hydrologic problems for their respective watersheds.

Understanding potential sources, and the geochemical variability and availability of major/trace elements becomes increasingly more important as populations increase and land uses change across the southwestern US.

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Table 1 Snowpack dust sample information. Sample numbers represent their corresponding dust storm event.

Sample ID	Zone	E	N	Dust Event	Date Collected	Leach step 1	Leach step 2	Leach step 3
1-CW	12 T	447790	4493939	2/6/14	4/24/14	1M acetic acid	0.8 M nitric acid	none
2-CW	12 T	447790	4493939	3/17/14	4/24/14	1M acetic acid	0.8 M nitric acid	aqua regia
2-GBNP	11 S	732327	4320706	3/17/14	4/11/14	1M acetic acid	0.8 M nitric acid	aqua regia
2-NW-A	12 T	452835	4637243	3/17/14	3/24/14	1M acetic acid	0.8 M nitric acid	aqua regia
2-NW-B	12 T	451729	4644345	3/17/14	3/24/14	1M acetic acid	0.8 M nitric acid	aqua regia
2-NW-C	12 T	457892	4634870	3/17/14	3/24/14	1M acetic acid	0.8 M nitric acid	aqua regia
2-SW-A	12 T	445356	4470897	3/17/14	3/29/14	1M acetic acid	0.8 M nitric acid	aqua regia
2-SW-B	12 T	448786	4472578	3/17/14	3/29/14	1M acetic acid	0.8 M nitric acid	aqua regia
2-WU	12 T	504379	4503049	3/17/14	3/25/14	1M acetic acid	0.8 M nitric acid	none
3-CW	12 T	447790	4493939	4/22/14	4/24/14	1M acetic acid	0.8 M nitric acid	none
3-WU-A	12 T	507089	4504010	4/22/14	4/24/14	1M acetic acid	0.8 M nitric acid	aqua regia
3-WU-B	12 T	502643	4503861	4/22/14	4/24/14	1M acetic acid	0.8 M nitric acid	none
4-CW	12 T	447790	4493939	4/5/15	4/11/15	1M acetic acid	0.8 M nitric acid	none
5-CW	12 T	504379	4503049	4/14/15	4/16/15	1M acetic acid	0.8 M nitric acid	aqua regia
5-WU	12 T	447790	4493939	4/14/15	4/21/15	1M acetic acid	0.8 M nitric acid	aqua regia

Table 2 Dust bucket sample information. Note that all samples represent an accumulation of dust that may have been deposited during the time the dust trap was deployed.

Sample ID	Zone	E	N	Date Deployed	Date Collected	Pre-treatment for XRD
SW1-2013	12 T	454578	4478954	9/11/13	10/20/13	30% hydrogen peroxide
WU1-2013	12 T	507171	4504571	9/18/13	10/18/13	30% hydrogen peroxide
WU2-2013	12 T	507171	4504571	9/18/13	10/18/13	30% hydrogen peroxide
WU3-2013	12 T	502247	4503021	9/18/13	10/18/13	30% hydrogen peroxide
WU4-2013	12 T	491335	4495370	9/18/13	10/13/13	30% hydrogen peroxide
SW2-2013	12 T	447356	4460396	9/11/13	10/31/13	30% hydrogen peroxide
GBNP1-2014	11 S	732936	4320608	9/28/13	7/1/14	30% hydrogen peroxide
GBNP2-2014	11 S	732195	4329515	9/28/13	7/1/14	30% hydrogen peroxide
GBNP3-2014	11 S	732936	4320608	7/1/14	9/1/14	30% hydrogen peroxide
GBNP4-2014	11 S	732195	4329515	7/1/14	9/1/14	30% hydrogen peroxide
WU5-2014	12 T	496313	4491481	7/15/14	10/31/14	30% hydrogen peroxide
SW3-2014	12 T	454371	4478689	7/15/14	10/31/14	30% hydrogen peroxide

WU6-2014	12 T	504379	4503049	7/15/14	10/31/14	30% hydrogen peroxide
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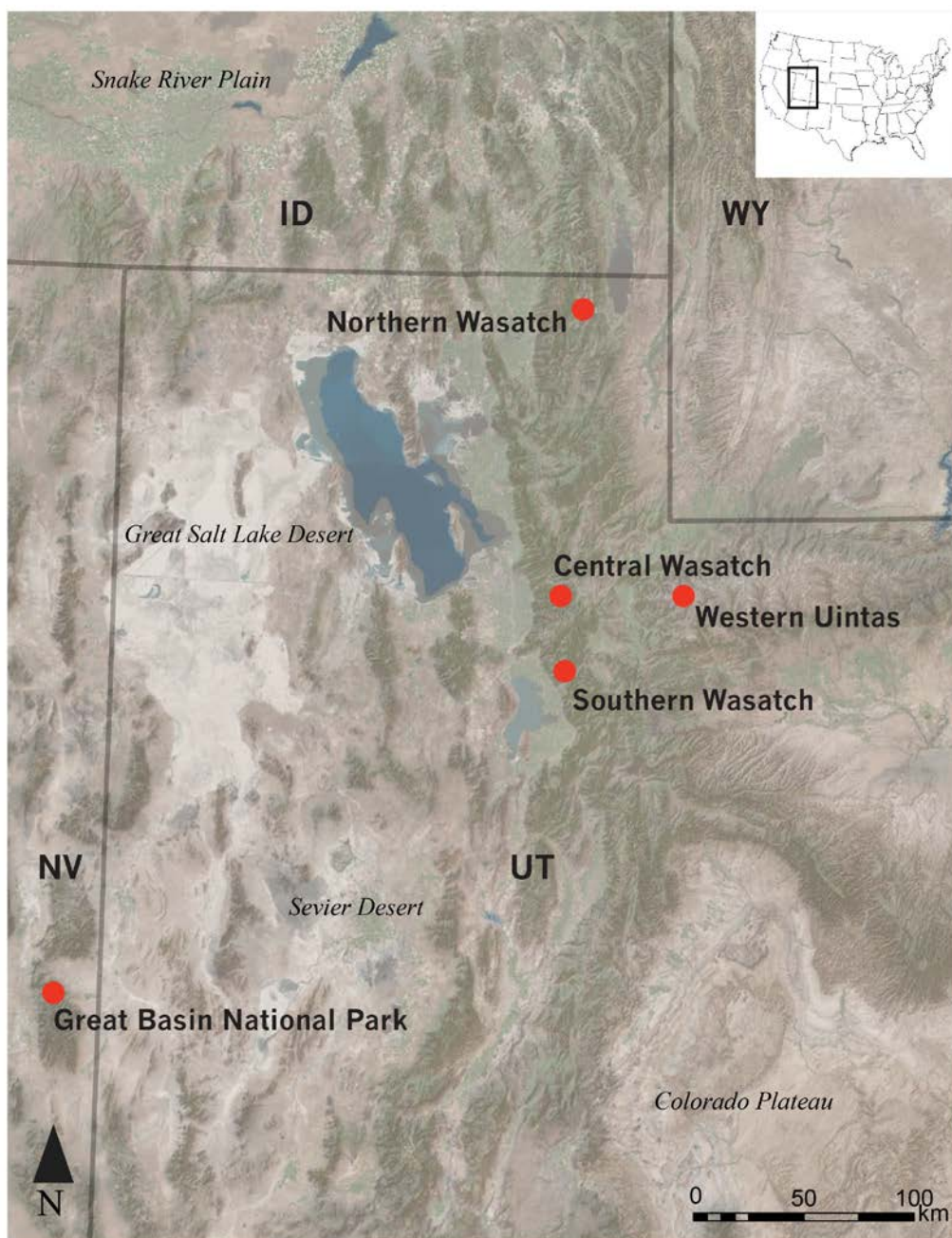


Figure 1 Location map showing sample areas (red dots) in Nevada (NV) and Utah (UT), USA. Regional dust sources are labeled in italics. Multiple dust samples were collected from each area during 2013 through 2015. See Table 1 for sampling details.

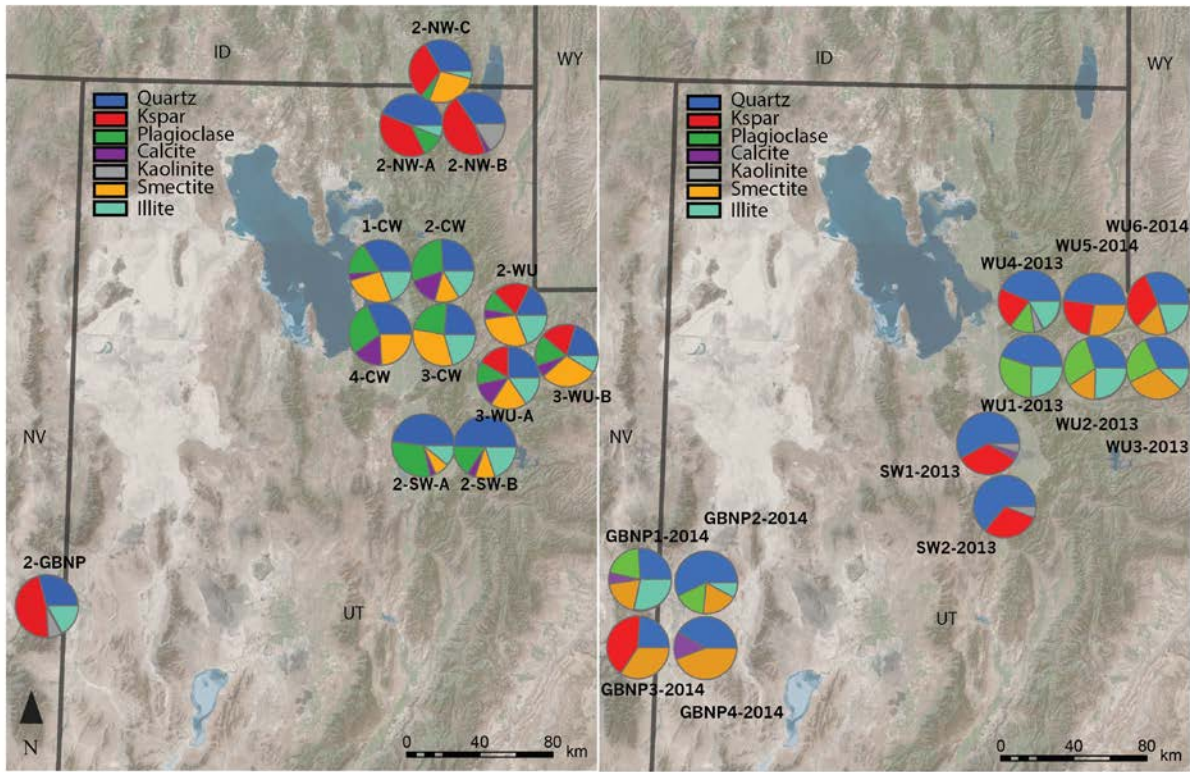


Figure 2 Pie charts showing mineralogy of snowpack dust samples (left side) and dust bucket samples (right side).

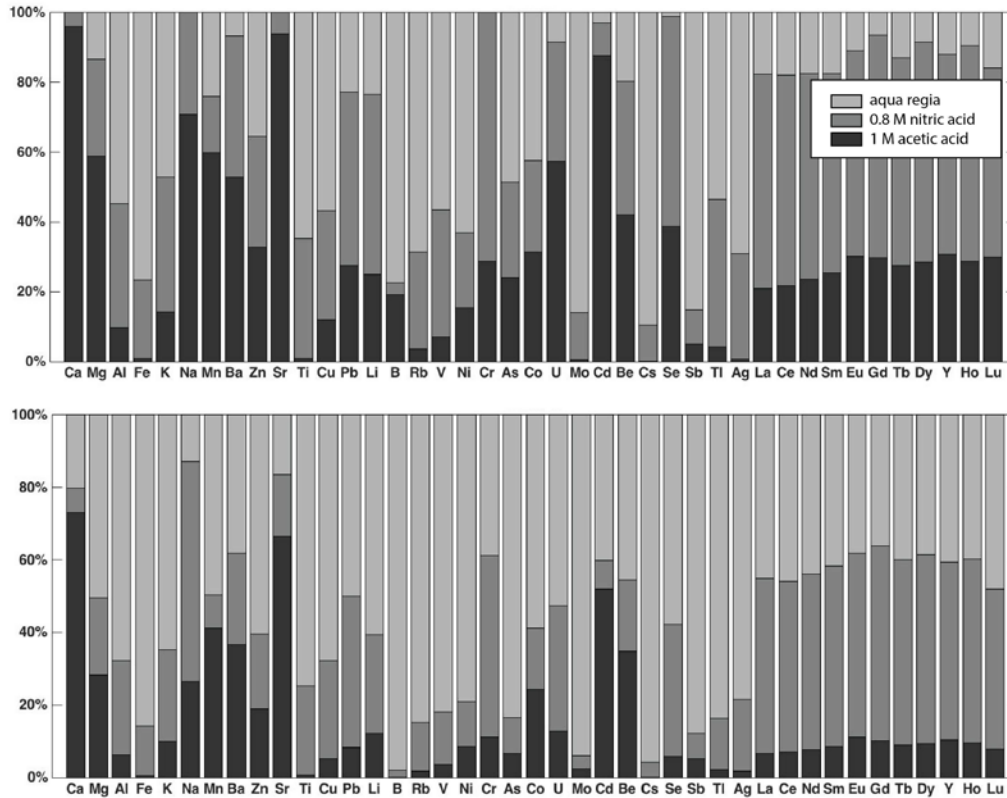


Figure 3 Relative fractions of selected trace and major elements and REE's in sequential leach steps using 1 M acetic acid, 0.8 M nitric acid, and aqua regia. Both samples are from the 17 March 2014, dust event. Top: central Wasatch Bottom: northern Wasatch. For all samples nearly 60 % of Ca, Sr and Cd were removed in the acetic acid leach compared with only 20 % of Mg, Na, Mn, Zn and Be. Cr and all REE had large organic components (~ 60 %). Because such a high percentage of Cr and all REE were removed with the nitric acid leach, it is possible these elements were associated with large organic particles in the dust samples (Lawrence et al., 2010). For the northern Wasatch and central Wasatch, other elements including Al, Fe, Ti, B, Rb, V, Ni, As, Mo, Cs Sb, Tl and Ag were found predominantly in the residual fraction (Lawrence et al., 2010).

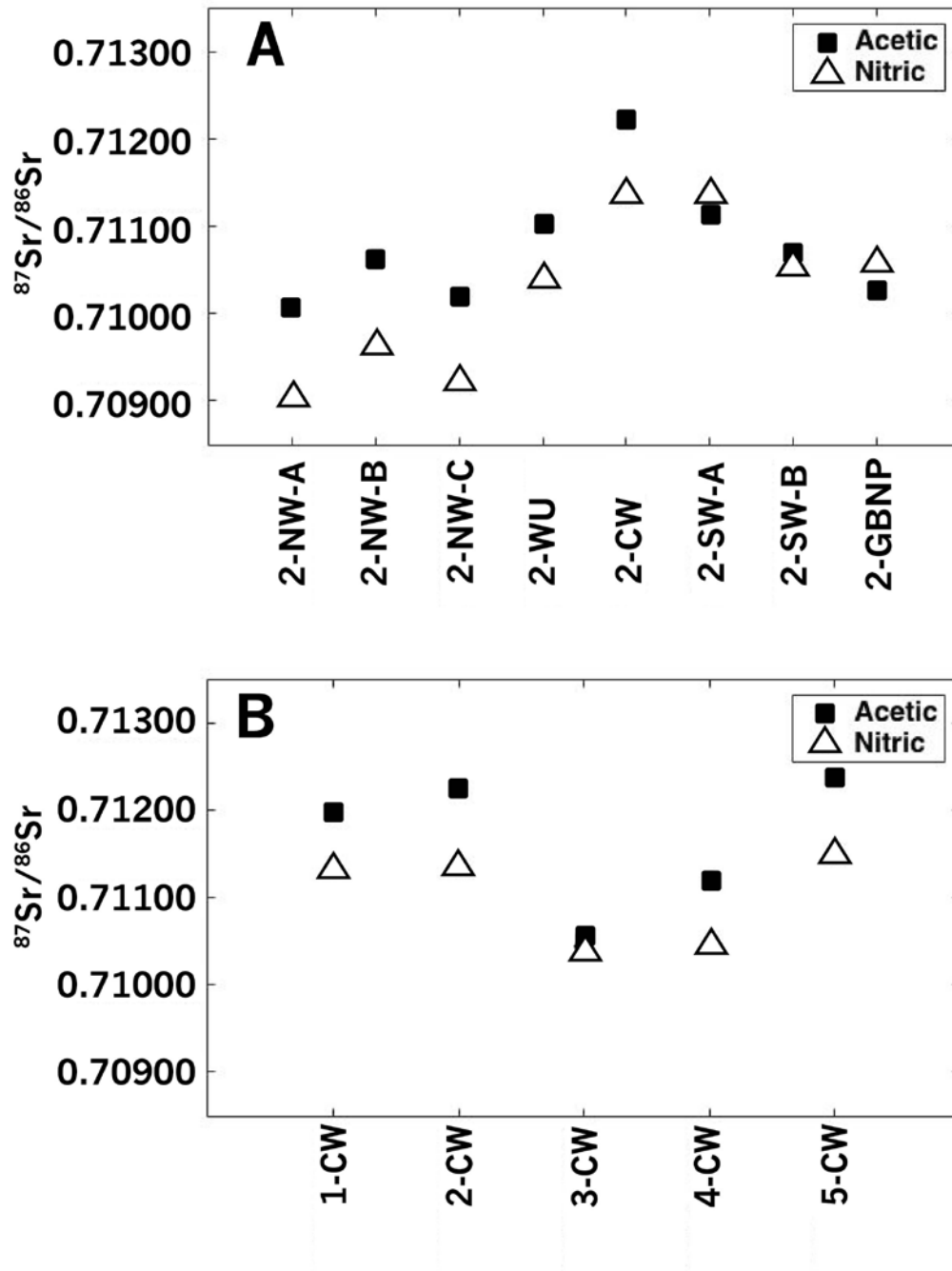


Figure 4 $^{87}\text{Sr}/^{86}\text{Sr}$ isotope ratios from snow dust samples from the 17 March 2014 dust storm event (top) and snow dust samples collected at the central Wasatch site (bottom).

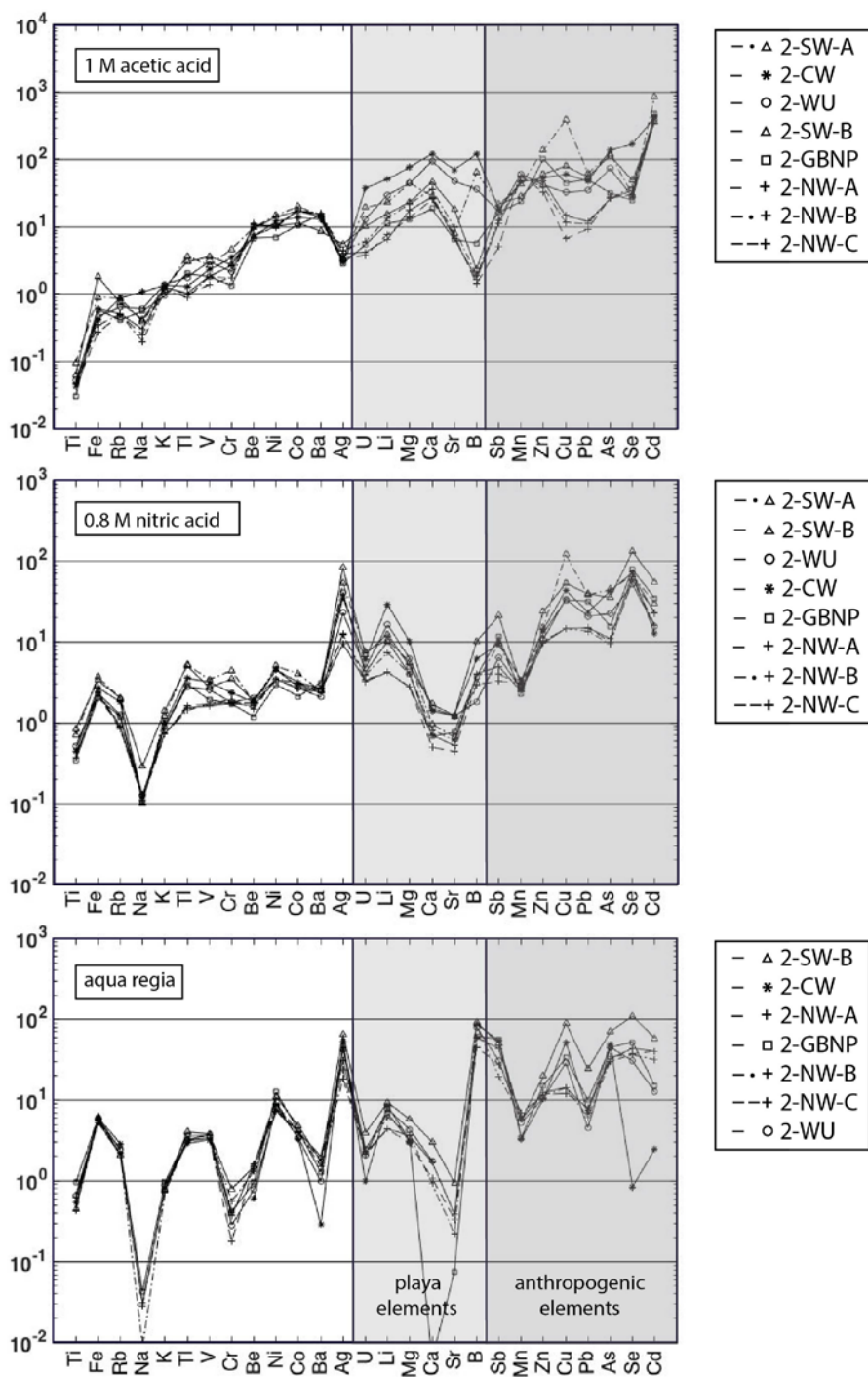


Figure 5 Major and trace element enrichment factors normalized to Aluminum in the average upper continental crust as reported by (Wedepohl, 1995) for all snow dust samples that captured the 17 March 2014 dust storm event. Note log scale.

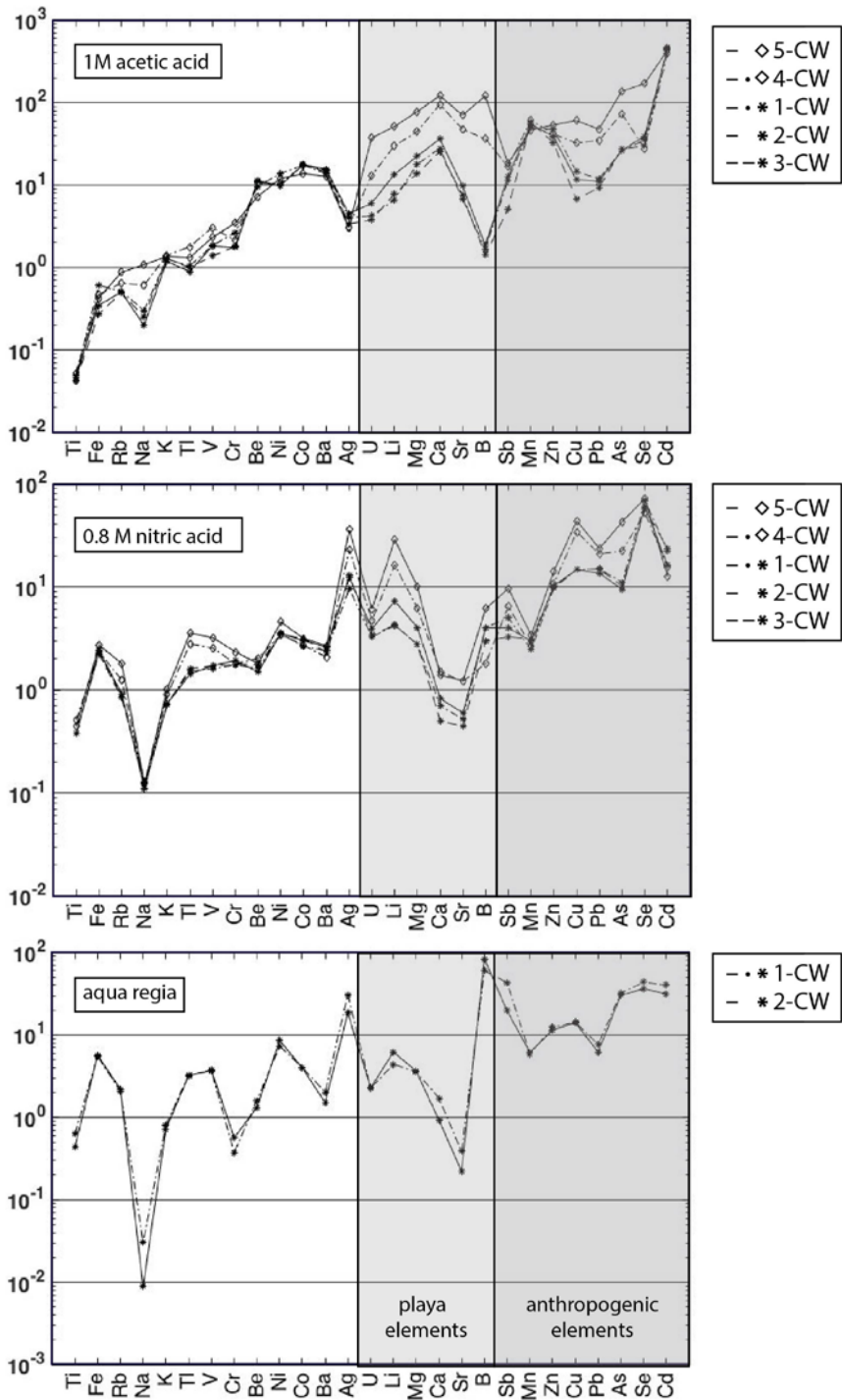


Figure 6 Major and trace element enrichment factors normalized to Aluminum in the average upper continental crust as reported by (Wedepohl, 1995) for all samples collected at the central Wasatch site showing five individual dust events over the span of two years.

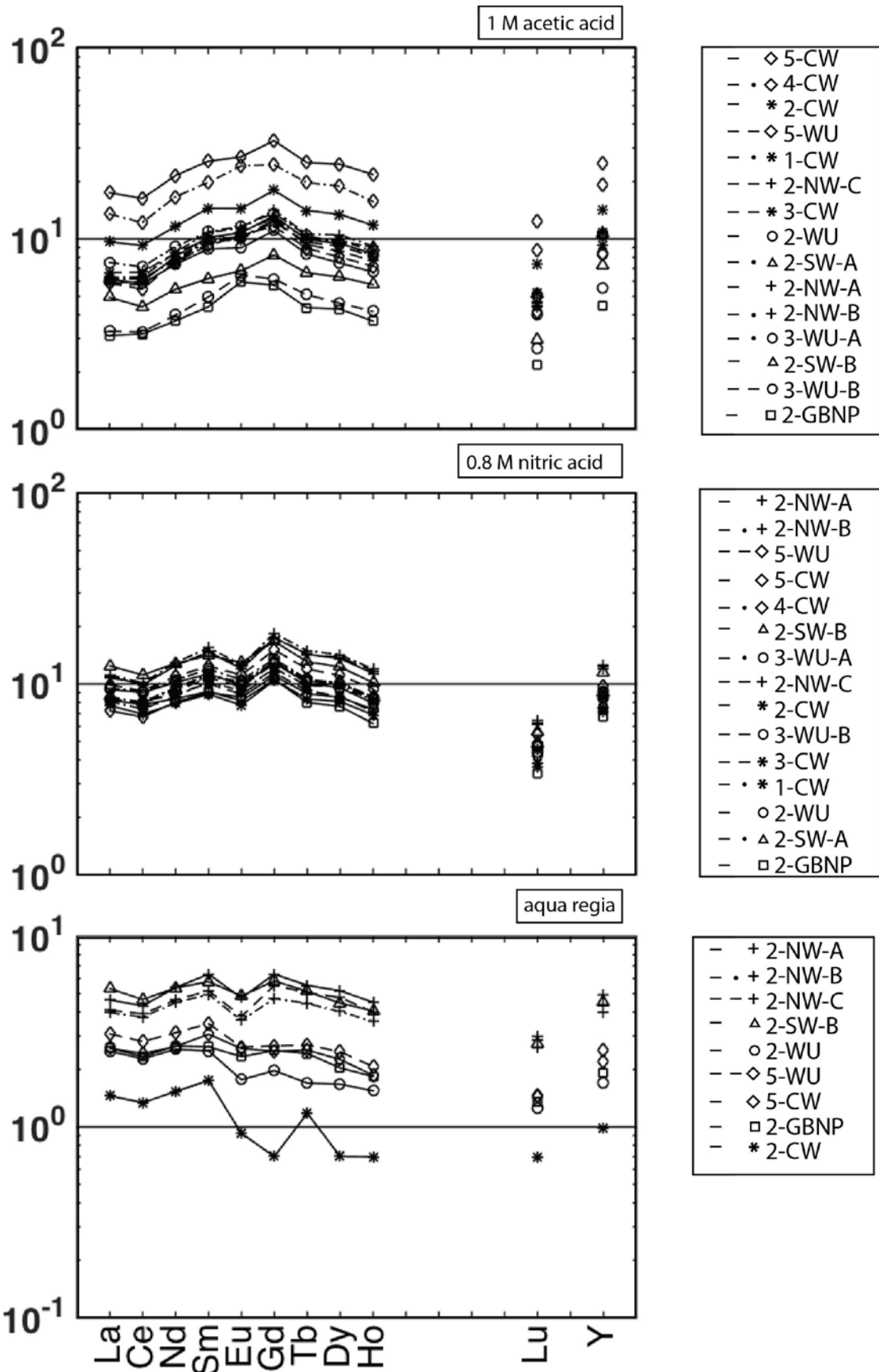


Figure 7 EFUCC for rare earth elements normalized to aluminum in the average upper continental crust as reported by (Piper, 2013). All snow dust samples coincided with the average concentration REE from the upper continental crust. A small negative Eu anomaly was present in the organic and residual fractions of all samples, which indicate the dust is at least partially derived (ultimately) from sources that underwent feldspar fractionation. Note log scale.

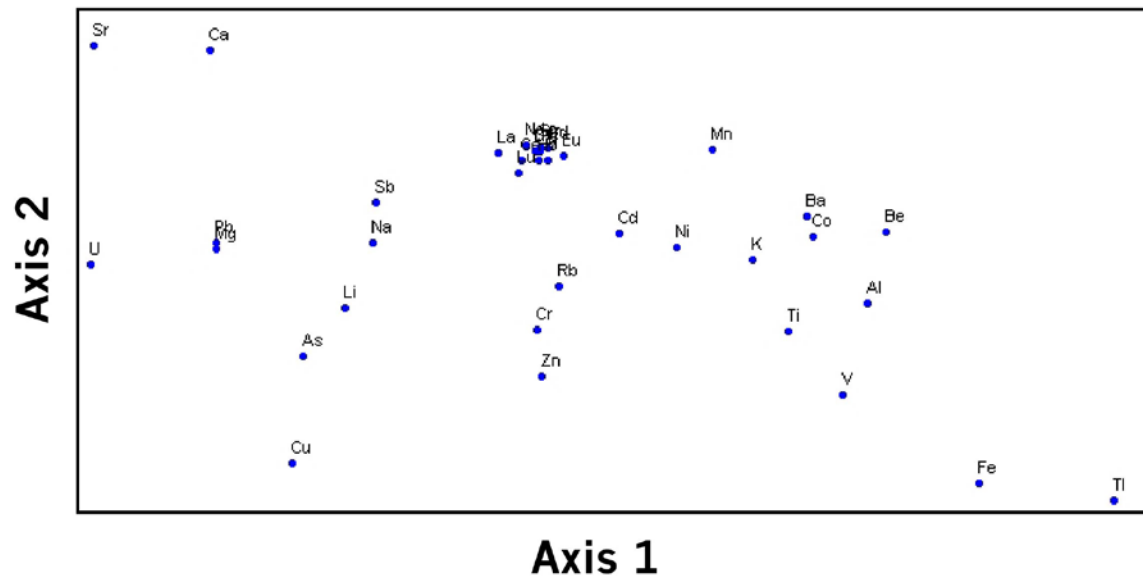
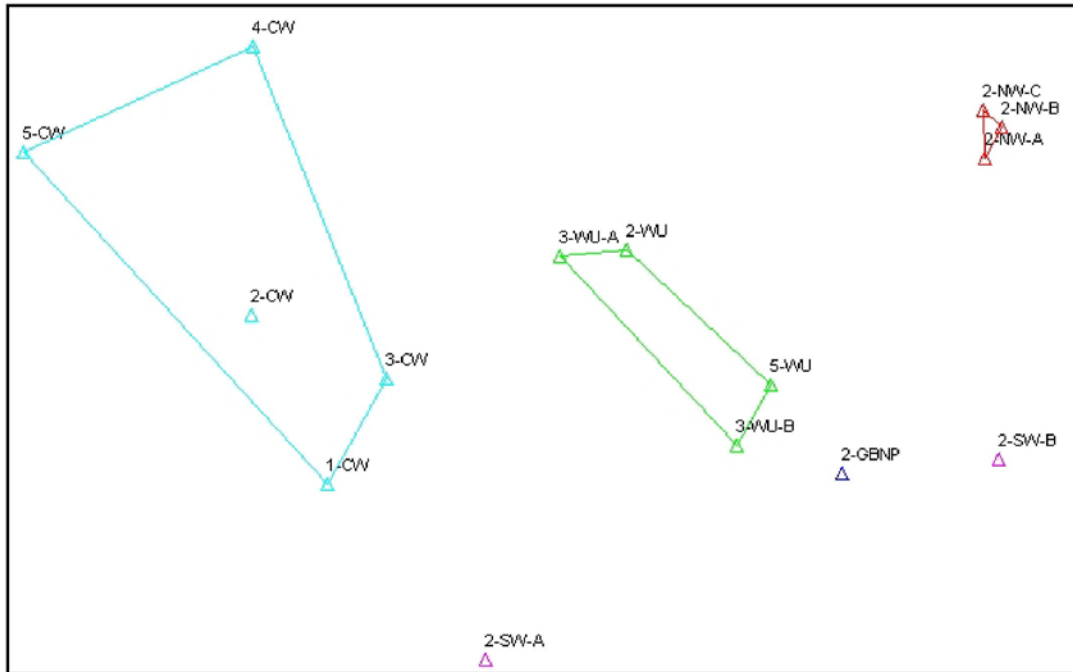


Figure 8 Non-metric Multidimensional Scaling MS for all snow dust samples. The data are best described using a two dimensional NMS model (final stress = 5.71). Peck (2010) states a final stress < 10 is acceptable. Axis 1 describes 80 % of the variability in the data and axis 2 describes 15 %. Sample locations have been grouped using polygons called “convex hulls”. Samples collected in the same sample location over the course of this study all plot near one another but sample locations plot in different regions of the ordination space. The northern Wasatch samples are defined by Tl and Fe. The central Wasatch samples are defined by elements associated with carbonate minerals (Ca, Sr, Mg) and heavy metals (U, Cu, Pb). The western Uintas samples plot in the middle of axis 1 and are primarily defined by trace elements (Ba, Co, Ti, Ni and Cd)

NOAA HYSPLIT MODEL
 Backward trajectories ending at 0300 UTC 18 Mar 14
 NARR Meteorological Data

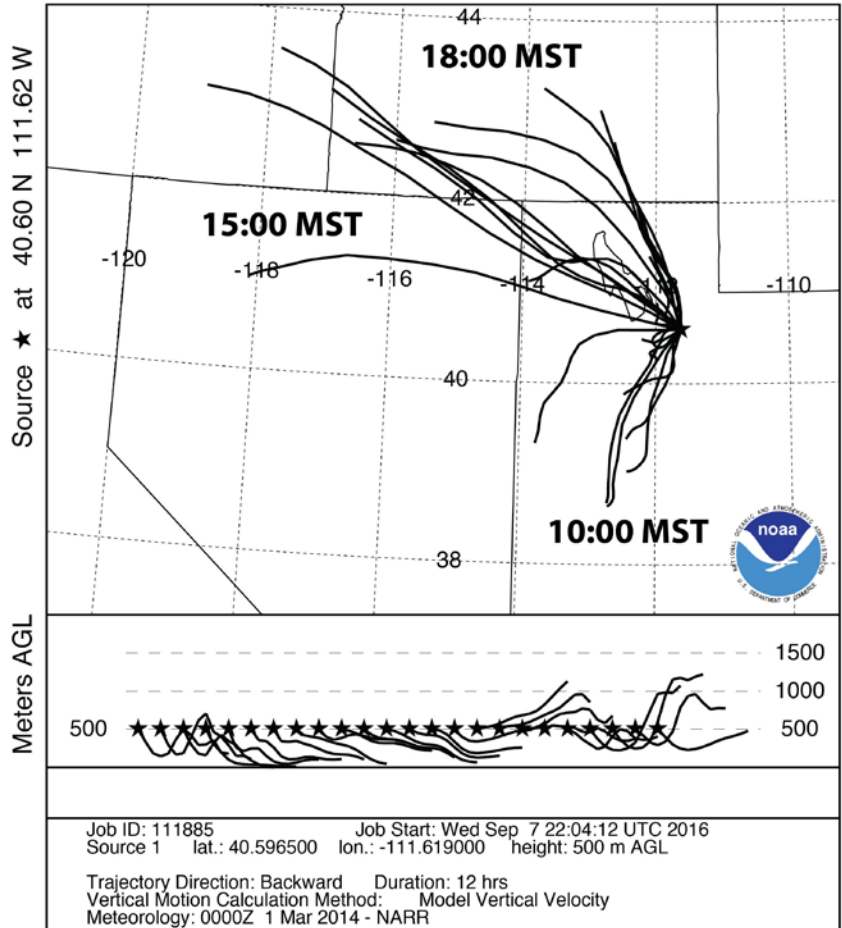


Figure 9 HYSPLIT back trajectories for air masses associated with the 17 March 2014 dust deposition event. Pre-frontal winds (~ 8 m/s) came from the south-southwest, increased in intensity (~ 16 m/s) and shifted to the west-northwest as the storm passed. Because there was such little precipitation, dust sources remained dry and were able to produce dust from both pre and post-frontal winds.