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# Geochemical characterization of critical dust source regions in the American West

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**Abstract**

The generation, transport, and deposition of mineral dust are detectable in paleoclimate records from land, ocean, and ice, providing valuable insight into earth surface conditions and cycles on a range of timescales. Dust deposited in marine and terrestrial ecosystems can provide critical nutrients to nutrient-limited ecosystems, and variations in dust provenance can indicate changes in dust production, sources and transport pathways as a function of climate variability and land use change. Thus, temporal changes in locations of dust source areas and transport pathways have implications for understanding interactions between mineral dust, global climate, and biogeochemical cycles. This work characterizes dust from areas in the American West known for dust events and/or affected by increasing human settlement and livestock grazing during the last 150 years. Dust generation and uplift from these dust source areas depends on climate and land use practices, and the relative contribution of dust has likely changed since the expansion of industrialization and agriculture into the western United States. We present elemental and isotopic analysis of 28 potential dust source area samples analyzed using Thermal Ionization Mass Spectrometry (TIMS) for $^{87}Sr/^{86}Sr$ and $^{143}Nd/^{144}Nd$ composition and Multi-Collector Inductively Coupled Plasma Mass Spectrometer (MC-ICPMS) for $^{176}Hf/^{177}Hf$ composition, and ICPMS for major and trace element concentrations. We find significant variability in the Sr, Nd, and Hf isotope compositions of potential source areas of dust throughout western North America, ranging from $^{87}Sr/^{86}Sr = 0.703699$ to 0.740236, $\varepsilon_{Nd} = -26.6$ to 2.4, and $\varepsilon_{Hf} = -21.7$ to $-0.1$. We also report differences in the trace metal and phosphorus concentrations in the geologic provinces sampled. This research provides an important resource for the geochemical tracing of dust sources and sinks in western North America, and will aid in modeling the biogeochemical impacts of increased dust generation and deposition caused by higher drought frequency and human activity.


*Keywords:* Aeolian dust; Sediment; American West; Strontium; Neodymium; Hafnium; Phosphorus

## 1. INTRODUCTION

Mineral dust is a critical component of Earth surface processes, and influences the present-day climate through a variety of direct and indirect means. In the atmosphere, dust modifies the Earth's radiative balance and cloud properties leading to the cooling or warming of surface temperatures (Mahowald, 2011; Mahowald et al., 2006). On snow and ice surfaces, the deposition of dust can decrease albedo

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and accelerate melt (Conway et al., 1996; Painter et al., 2007). Dust is also an important constituent within continental and marine biogeochemical cycles; it provides biologically limiting nutrients (including Fe, Si, and P) to the oceans, thereby increasing biological productivity and the uptake of atmospheric $CO_2$ in high nutrient, low chlorophyll regions (Jickells et al., 2005). These same nutrients are also provided by atmospheric deposition to continental ecosystems, leading to ecological sustenance in nutrient-limited landscapes (Prospero, 1999; Shinn et al., 2000; Aciego et al., 2017). Conversely, in nutrient-rich ecosystems, dust deposition may result in eutrophication, which can degrade water quality, and, in the case of trace metals, lead to soil toxicity (Sickman et al., 2003; Ballantyne et al., 2011).

One of the reasons mineral aerosols have become an important topic in Earth system sciences is that their deposition, and impact, leave detectable traces in paleoclimate records from land, sea, and ice (Biscaye et al., 1997; deMenocal et al., 2000; Fischer et al., 2007; Kohfeld and Harrison, 2003; Rea, 1994; Sugden et al., 2009; Winckler et al., 2008; Winckler and Fischer, 2006, and others), providing insight into Earth surface conditions on variable timescales. Dust production is diverse across the Earth's surface, and source regions occur in a variety of environments (Prospero et al., 2002), leading to variable nutrient concentrations and unique geochemical signatures indicating provenance. Past work by Biscaye et al. (1997), Kanayama et al. (2002), and Chen et al. (2007) have provided an abbreviated list of the isotopic compositions of dust from potential source areas (PSA) in the Northern Hemisphere; additional extensive mineralogical, chemical, and/or isotopic characterizations of site-specific locations in the desert southwest and Colorado Plateau have previously been made and catalogued (Ballantyne et al., 2011; Dahms, 1993; Lawrence et al., 2011; Neff et al., 2008; Reheis et al., 2009). Here, we focus on characterizing PSAs known for dust events and/or to have been affected by increasing western settlement and livestock grazing during the last 150 years.

Land cleared for agriculture and human settlement throughout the United States (US) from the time of initial European immigration has led to both direct and indirect impacts upon climate. Deforestation and the transformation of land into croplands and pastures have affected the terrestrial carbon cycle (Houghton et al., 1999), while increased human activity such as agriculture and livestock grazing in the American southwest and the Colorado Plateau during the past several decades has been correlated with greater dust production and deposition, and has been identified on mountain snow packs on both seasonal to annual timescales (Painter et al., 2007; Reynolds et al., 2001). Mountain snowpack records reveal that during the time period 1925–2000 AD, the American West (i.e. from the Continental Divide to the Pacific and from central British Columbia, Canada to southern Arizona and New Mexico) has experienced widespread declines in springtime snow water equivalent due to warming, and model simulations warn that it is the most susceptible to future warming (Mote et al., 2005). Natural climate variability and human forcing has been correlated to current and future water supply crises in the southwestern US using regional (Barnett et al., 2008) and global models (Latif and Barnett, 1994). Dust generation and uplift from potential source areas depends on climate and land use practices, and it is likely that the relative contributions to dust mass loadings has changed since the expansion of industrialization and agriculture into the western US. The impact and geochemical characterizations of such dust sources on these systems is discussed below.

### 1.1. Radiogenic isotopes and trace elements as indicators of dust provenance

Many studies have considered dust inputs to ecosystems in the western US (Aciego et al., 2017; Ballantyne et al., 2011; Brahney et al., 2013, 2014, 2015; Lawrence et al., 2011; Neff et al., 2008; Reheis, 1997; Reheis, 2006; Reheis et al., 1995, 2002a, 2009; Reheis and Kihl, 1995; Reheis and Urban, 2011; Reynolds et al., 2001, 2006a, 2006c, 2010), however less is known about the dust sources and pathways. The existing library of dust in the American West is limited (see Fig. S1), and a more broadly defined catalog of the chemical composition of dust sources in the western US is needed to provide better constraints on the variability (or similarity) of source material. In this study, we chose sites that were most likely to generate large amounts of dust based on existing dust cycle literature for the western US. Determining source provenance is possible using radiogenic isotope compositions (Sr, Nd, and Hf), as variations in geologic history and crustal ages of different source regions will impart distinctive isotope compositions (Biscaye et al., 1997; Grousset and Biscaye, 2005; Grousset et al., 1992). Measurements of the radiogenic isotopes of both neodymium ($^{143}Nd/^{144}Nd$) and strontium ($^{87}Sr/^{86}Sr$) are traditionally used together to distinguish lithological source areas and have proven to be an excellent "fingerprint" of dust source (Grousset and Biscaye, 2005). Combined Nd and hafnium (Hf) isotopic compositions ($^{176}Hf/^{177}Hf$) are likewise useful in determining the degree of continental weathering of earth materials (Bayon et al., 2006; Rickli et al., 2009, 2010; van de Flierdt et al., 2007) due to the incongruent weathering of Hf from zircons (Piotrowski et al., 2000; van de Flierdt et al., 2007). Hf isotope compositions of aerosol mineral dust in sinks, or depositional environments (i.e. ice sheets, glaciers and oceans) can be used to estimate provenance and transport distance (Aarons et al., 2013).

Trace element concentrations and ratios in modern dust deposits and the paleoclimate record also provide a mechanism for reconstructing dust provenance and nutrient input from exogenic sources (Chadwick et al., 1999; Feng et al., 2011; Hinkley and Matsumoto, 2001; Jahn et al., 2001; Marx et al., 2005; McGowan et al., 2005). In particular, trace heavy metals (i.e. Pb, As, Zn Cu, Cr, and Ni) in ice and sediment core records are known to be powerful indicators of air pollution from industrialization, such as fossil fuel combustion, smelting, and coal mining (McConnell and Edwards, 2008; McConnell et al., 2002; Reheis et al., 2009). Trace element concentration data for PSA samples

from the American West will provide additional constraints on the transport pathways of aerosols and the presence of anthropogenically sourced pollutants for existing and future paleoclimate records.

North American dust sources are poorly characterized in terms of their isotope compositions, and there is little existing data demonstrating the relationship between anthropogenic influence and climate variability upon dust sources. Previous work characterizing the Sr and Nd isotopic ratios of alpine lake core sediments, snowpack, and soils in the San Juan Mountains of Colorado has indicated that dust deposited in the San Juan Mountains is likely derived from a variety of regional sources (Ballantyne et al., 2011; Lawrence et al., 2011; Neff et al., 2008; Painter et al., 2007). An extensive transect across the dust deflation areas of the southwest US provided a narrow range in Sr isotopic compositions, although Nd isotopes were not measured (Goldstein et al., 2008). The Sr-Nd isotopic characterization of mineral dust has also been conducted on Asian aerosols (Kanayama et al., 2005) and PSA samples (Chen et al., 2007), and more recent studies have focused on characterizing the Nd and Hf isotope compositions of PSAs (Zhao et al., 2014).

### 1.2. Dust proxy measurements in the western US

Intensification of land use throughout the western US combined with increased drought frequency and stronger wind speeds can lead to greater dust availability, transport, and deposition (Ballantyne et al., 2011; Belnap and Gillette, 1997; Neff et al., 2008; Reheis and Urban, 2011). The natural and anthropogenic influences on the dust cycle in the western US have been explored in a number of studies using dust records preserved in sediment cores, mountain snowpack, and ice cores (Aarons et al., 2016a; Brahney et al., 2013, 2014, 2015; Doebbert et al., 2014; Neff et al., 2008). For example, Neff et al. (2008) showed that recent dust storms incident on the San Juan Mountains of Colorado (and associated alpine lakes) have been dominated by large dust particles (>37 μm). Particles in the silt size class (2–63 μm) are common in western US dust storms, and there is increasing evidence that these particles can travel hundreds of kilometers from their source (Neff et al., 2013). Further work on dust events in the western US using a combination of lake records, snow pack, and back trajectories in the San Juan Mountains of Colorado suggest that the dust was primarily sourced from the Great Basin, the Colorado Plateau, Mojave and Sonoran Deserts (Neff et al., 2008; Painter et al., 2007). It has been found that the impact of agricultural development and water diversion also results in higher local dust emissions from the Colorado Plateau and Green River Basin regions (Brahney et al., 2013; Brahney et al., 2014; Doebbert et al., 2014; Neff et al., 2008; Painter et al., 2007). Furthermore, the biogeochemical impact of increased dust deposition to mountain lakes has become a topic of interest (Brahney et al., 2014; Brahney et al., 2015; Neff et al., 2008; Reynolds et al., 2010), with increased nutrient and heavy metal concentrations having discernable biological impacts on lake ecosystems (Brahney et al., 2015, 2014).

Variations in dust provenance can indicate changes in aridity, dust production, and atmospheric transport pathways (Delmonte et al., 2004; Fischer et al., 2007; Rea, 1994; Wolff et al., 2006); however, determining the sources and pathways of atmospheric mineral dust and marine aerosols remains largely unconstrained. In order to address this problem, researchers have investigated combinations of mineralogy, elemental compositions, radiogenic isotope compositions, and size distributions of both PSAs and dust deposited in ice from Europe, Asia, Antarctica, and Greenland (Aarons et al., 2016b; Blakowski et al., 2016; Delmonte et al., 2004; Lupker et al., 2010; Thevenon et al., 2009).

In this work, we utilize measurements of strontium (Sr), neodymium (Nd), and hafnium (Hf) isotope compositions, along with trace element concentrations in samples from PSAs in the western US to geochemically fingerprint each source region. The data presented here serves as a chemical and isotopic catalogue of dust sources throughout arid regions in North America for future use in disentangling the dust sources and pathways preserved in paleoclimate records.

## 2. STUDY SETTING

This report describes an investigation of surficial sediment in a number of desert regions throughout the American West (Fig. 1) conducted to provide greater constraints on the variability (or similarity) of source material chemistry based on surrounding geology and proximity to industrialization and cultivated land. To focus our collection efforts across a vast study area, we established sampling localities covering an array of landforms, elevations, bedrock, flora, weather patterns, and land-use histories. For the purposes of comparing and contrasting the numerous data, the 28 total sample sites have been separated into five

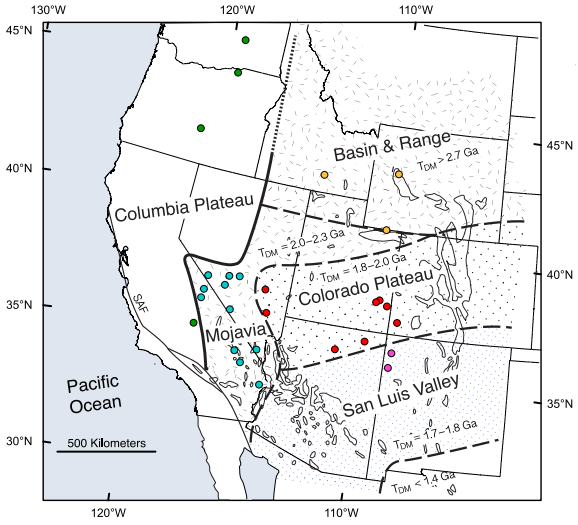


Fig. 1. Location map of PSA sampling sites (indicated by filled circles) for this study, divided into the five crust-formation provinces of the western US as outlined by Bennett and DePaolo (1987) via neodymium isotopic mapping.

regions based on a map of crust-formation provinces of the western US constructed by Bennett and DePaolo (1987), wherein boundaries are constrained by regions of similar Nd isotopic compositions. These regions are described in detail below, while information on individual dust collection sites is provided in Table 1.

### 2.1. Columbia plateau

The Columbia Plateau is a geologic province covering portions of Washington (WA), Oregon (OR), and Idaho (ID) located between the Cascade Range and the Rocky Mountains. In this study, we include a sample from Goodale Creek, California (CA) (N 35°58.789′, W 118°16.482′, elevation: 1245 m.a.s.l.) in the Columbia Plateau region, as this falls within the same crust-formation province as the remaining samples from WA and OR. During the Miocene, an extended period of powerful volcanism engulfed the Columbia Plateau with thick layers of flat-lying flood-basalt flows (Tolan et al., 2009). In more recent geologic time, glacial outburst flood sediments subsequently reworked by wind have formed large deposits of silt, clay, and fine sand, referred to as loess (Claiborn et al., 1998; McDonald and Busacca, 1992; Sweeney et al., 2005). Due to the rain shadow cast by the Cascade Range, the present-day regional climate is arid to semi-arid (McDonald and Busacca, 1992); the sandy, gravelly soils support vegetation dominated by shrubs such as sagebrush and bitterbrush, as well as a variety of perennial grasses (Sweeney et al., 2005). In addition to the "loose" compositional nature of aeolian loess deposits, lack of soil organic material, and dry climatic conditions, the erodibility of surface materials is further enhanced by heavy agricultural land use, particularly after the fall harvest when both irrigated and fallow fields are left bare (Claiborn et al., 1998).

The northernmost PSA investigated during this study was Moses Lake Sand Dunes, WA (N 47°04.018′, W 119° 17.254′, elevation: 347 m.a.s.l.), which is the largest dune-

Table 1
List of field sites for collection of dust source material throughout the American West.

| Sample site | Sample name | Latitude (N) | Longitude (W) | Elevation (m.a.s.l.) | Local substrate | Source type |
|---|---|---|---|---|---|---|
| *Basin and range* | | | | | | |
| Green River Basin, UT | GRB-060514 | 40.9798 | 109.8366 | 2200 | Shale/sandstone | Alluvium |
| Snake River Plain, ID | SRP-060514 | 42.6795 | 113.4848 | 1295 | Rhyolite/alkalic basalt sediments | Sand dunes |
| DuBois, WY | R411 | 43.4251 | 109.5677 | 1250 | Granite | Glacial moraine |
| *Colorado plateau* | | | | | | |
| Alamosa Valley, CO | AV-060814 | 37.4742 | 105.6409 | 2324 | Sandstone | Alluvium |
| Chilchinbito Region, AZ | CHI-061114 | 36.5836 | 110.0611 | 1700 | Sandstone/Sediments | Sand dunes |
| Cadiz Lake, NV | CL-061414 | 37.8584 | 115.2533 | 1508 | Rhyolite/granite | Dry playa |
| Coyote Springs, NV | CS-061414 | 36.9886 | 114.9783 | 763 | Limestone | Sand dunes |
| Dugout Ranch, UT | DR-060614 | 38.0559 | 109.2526 | 1676 | Mudstone/siltstone/sandstone | Alluvium |
| Little Colorado Corridor, AZ | LCC-061114 | 36.0938 | 111.4138 | 1437 | Sandstone/Sediments | Alluvium |
| Needles, UT | NDL-060614 | 38.2500 | 109.6513 | 1917 | Shale | Shallow sea deposit |
| Virginia Park, UT | VP-060714 | 38.1523 | 109.8054 | 525 | Sandstone | Shallow sea deposit |
| *Columbia plateau* | | | | | | |
| Christmas Lake Valley, OR | CLD-060314A, B | 43.3462 | 120.3885 | 1330 | Basalt | Sand dunes |
| Goodale Creek, CA | GC-061516 | 35.9798 | 118.2747 | 1245 | Rhyolite/basalt | Alluvium |
| Moses Lake Sand Dunes, WA | MLD-060214 | 47.0669 | 119.2875 | 347 | Basalt | Sand dunes |
| Stanfield, OR | SF-060214 | 45.7865 | 119.2306 | 525 | Basalt | Alluvium |
| *Mojavia* | | | | | | |
| Bare Mountain, NV | BM-061414 | 38.1448 | 116.6194 | 357 | Limestone | Alluvium |
| Crater Flat, NV | CF-061514 | 36.8132 | 116.7268 | 900 | Metamorphic/mixed | Alluvial fan |
| Goldfield, NV | GF-061515 | 37.6962 | 117.2367 | 1781 | Basalt | Dry playa |
| Kelso Dunes, CA | KD-061314 | 34.8948 | 115.6409 | 796 | Rhyolite/granite | Sand dunes |
| McCullough Range, NV | MR-061314 | 35.5118 | 115.0733 | 1314 | Rhyolite/granite | Alluvium |
| Upper Silver Lake, CA | SL-061314 | 35.3058 | 116.0869 | 272 | Rhyolite/granite | Dry playa |
| Stooge Range, CA | STR-061514 | 37.0014 | 118.2152 | 1177 | Basalt | Alluvial fan |
| Trail Canyon, NV | TC-061614 | 37.9019 | 118.1828 | 1574 | Basalt | Alluvium |
| Turtle Mountains, CA | TM-061214 | 34.2085 | 114.6176 | 309 | Metamorphic/mixed | Alluvium |
| Tonopah, NV | TON-061414 | 38.0720 | 117.1209 | 1675 | Rhyolite/granite | Alluvium |
| White Mountains, CA | WM-061614 | 37.3516 | 118.1828 | 2712 | Metamorphic/mixed | Alluvium |
| *San Luis valley* | | | | | | |
| Newcomb Region, NM | NR-061114 | 36.2793 | 108.7047 | 1695 | Sandstone/Sediments | Alluvium |
| Twin Lakes Region, NM | TLR-061114 | 35.7013 | 108.7744 | 1954 | Sandstone/Sediments | Alluvium |

field in the state. Similar to the majority of dune landforms found in central and southeastern WA, the dunes at Moses Lake consist mainly of quartz, with at least minor traces of basalt (Edgett and Lancaster, 1993). Our sample was taken from a lightly vegetated, active dune. Further south, we obtained a sample from a lower elevation (173 m.a.s.l.) outside a sandy terrace near Stanfield, OR, (N 45°47.192′, W 119°13.840′). This sample was comprised of similar material and surrounded by dry grasses and brush.

In the southeastern portion of OR, we sampled a PSA previously investigated by Edgett and Lancaster (1993), which is referred to as the Christmas Lake Valley Dunes (N 43°20.770′, W 120°23.310′). Two samples were collected here, the first from the crest at 1330 m.a.s.l., which we noted had a thin mineral crust, and the second from approximately 30 m below, which consisted of a finer material protected under a thicker, white mineral crust. When the Pleistocene lakes of the North American Great Basin dried up, lake sediments were frequently blown eastward to form elongate and transverse dunes, which previous studies have indicated are mostly composed of alkali feldspar and quartz (Edgett and Lancaster, 1993; Walker, 1977). Dole (1942) reported the mineralogy of a sample collected from this same region as 50% feldspar and 50% pumice.

### 2.2. Basin and range

The two PSAs sampled in ID and Wyoming (WY) are grouped according to their geochemically-distinct Archean crust (Mueller and Frost, 2006). The first is the dusty, eastern portion of the Snake River Plain (SRP) (N 42°40.767′, W 113°29.089′), a basaltic graben covered by soils formed from the fine materials carried by the Late Pleistocene Bonneville Flood (Hughes et al., 1999; Malde, 1968). Bounded to the north and south by mountains and valleys (or horsts and grabens) associated with the Basin and Range province, the fine-grained soils of the SRP have been agriculturally developed for the production of potatoes, grains, and sugar beets (Hughes et al., 1999). Here, the representative sample was collected at an elevation of 1295 m.a.s.l. from a sage-covered dune, and consists of a highly weathered mixture of sediments possibly derived from rhyolite and basalt.

The remaining WY samples come from the Wind River Range (WRR), a northwest-southeast trending mountain range with a core of Archean rocks (mainly felsic gneisses and granitoids), bounded on its east, south, and west by the semi-arid Wind River, Great Divide, and Green River Basins, all three of which have mean elevations greater than 2100 m.a.s.l. (Dahms, 1993; Stuckless et al., 1985). To the west, the Wind River thrust fault separates the Wind River massif from the much younger Eocene deposits of Utah's Green River Basin (Stuckless et al., 1985). There are no unmetamorphosed volcanic rocks in the WRR (Dahms, 1993). The WRR has been extensively glaciated throughout the Quaternary, and currently contains upwards of 40 alpine glaciers, making it the highest concentration of glaciers in the American Rocky Mountain system (Cheesbrough et al., 2009; Graf, 1977). However, temperature increases over the past century have led to a nearly continuous retreat of smaller alpine glaciers (Dyurgerov and Meier, 2000); using Landsat data, Cheesbrough et al. (2009) estimated that glacier cover in the WRR has decreased by over 25% since 1985, thereby exposing large quantities of fine-ground, unconsolidated and unsorted particulate material available for aeolian deflation and transport. Due to the increased sediment availability, many geochemical studies of soil and aeolian sediments have been conducted in the WRR (Blum and Erel, 1997; Dahms, 1993; Dahms and Rawline, 1996; Gosse et al., 1995; Hall, 1999; Harlavan et al., 2009). One of our WRR PSA samples was collected from a road cut along Forest Service Road 411, approximately 6.4 km from the trailhead for Glacier Trail on the east side of the range (N 43°25.51, W 109° 34.77, elevation: 1250 m.a.s.l.). This sample was collected from a past terminal moraine, comprised of mixed sedimentary and poorly sorted metamorphic rocks. The second WRR PSA sample is from the Big Meadow Dune on the Glacier Trail (N 43°15.715′, W 109°34.167′, elevation: 2947 m.a.s.l.), a U-shaped valley consisting of loess sand dunes. Both surface materials were covered by a thin crust and were surrounded by sagebrush and pine trees. The third and final WRR PSA sample was collected from a joint terminal moraine of the Gannett and Dinwoody glaciers (N 43°11.049′, W 109°37.609′, elevation: 3264 m.a.s.l.), and consists of poorly sorted granite and gneiss.

The Green River Basin (GRB) lies southwest of the WRR, comprised of silt-rich sediments that are exposed to wind erosion much of the year. These sediments contain volcanic minerals that are unique to the middle and late Eocene sedimentary formation in this region (Dahms, 1993). The Wasatch Formation is exposed only in the northern GRB, and is primarily red and gray shale, siltstone, and gray and brown sandstone (Cashion, 1967), whereas the Green River Formation is comprised of beds of oil shale, marlstone, shale, siltstone, sandstone, limestone and tuff deposited in a lacustrine environment (Cashion, 1967). The GRB region offers a source for windborne silt because it contains only scattered salt-tolerant shrubs as a cover against deflation. The PSA sample collected from the GRB (N 40°58.728′, W 109°50.196′, elevation: 2200 m.a.s.l.) was surrounded by abundant dry vegetation.

### 2.3. Colorado plateau

The Colorado Plateau is a high-elevation region consisting of Paleozoic and Mesozoic sedimentary rocks divided by broad open valleys and deeply incised canyons (Hunt, 1956). It covers more than 380,000 square km in southeastern Utah (UT), northern Arizona (AZ), northwestern New Mexico (NM), and southwestern Colorado (CO). Here, the climate is semiarid and erosion is at a maximum during arid climate periods (Hunt, 1956), suggesting that this area is particularly susceptible to increased dust generation with drought. In this study, the area sampled encompasses a portion of southwestern CO, southeastern UT, and southeastern Nevada (NV) (Fig. 1). The central Colorado Plateau in particular is a noted area of dust accumulation (Reheis et al., 2005; Reynolds et al., 2006c), possibly due to dust plumes from the central Mojave Desert that are driven east

by west-southwesterly winds (Reynolds et al., 2003). Further geochemical studies have indicated that the fine-grained sediment present on isolated surfaces on the Colorado Plateau originates from a distant source beyond the Colorado Plateau as well as local sources (Goldstein et al., 2008).

The first group of PSA samples from the Colorado Plateau were collected from three semi-arid, high-elevation (~1500–1900 m.a.s.l.) sites within the Needles district of Canyonlands National Park, located ~100 km south of Moab, UT, in the central area of the Colorado Plateau. For all sites, the sampled substrates consist of material weathered from the Cedar Mesa Sandstone. The region is characterized by nearly flat-lying Paleozoic and Mesozoic sandstone, silty sandstone, and arkosic beds (Reynolds et al., 2006c). Much of the vegetated landscape consists of aprons of sandy surficial deposits that slope gently away from bedrock exposures (Reynolds et al., 2006c).

We sampled from: (1) Dugout Ranch (N 38°03.357′, W 109°15.156′, elevation: 1676 m.a.s.l); (2) Virginia Park (N 38°09.140′, W 109°48.326′, elevation: 1553 m.a.s.l.), a sheltered basin of stabilized sand dunes overlying the Mesa Formation (Billingsley et al., 2002; Reheis et al., 2005); and (3) the Needles area (N 38°15.001′, W 109°39.078′, elevation: 1917 m.a.s.l.), an open broad basin surrounded by mesas and sparse desert vegetation. Previous work at Virginia Park has identified the uppermost sediment (0–10 cm) as derived primarily from sandstone bedrock (Reynolds et al., 2006c). It has been noted that the size of sediment particles at Virginia Park is dependent upon the geomorphic setting and slope of the sample site (Reynolds et al., 2006a). The Needles area is located in Canyonlands National Park within Virginia Park. The area is a basin consisting of vegetated sand dunes that are bordered by sandstone cliffs, and small drainage networks intersect the Quaternary sediment. Hack (1941) referred to the Four Corners region (including Virginia Park and the Needles area) of the Colorado Plateau as the largest area of aeolian sand in the southwestern US. The dune sands in the Four Corners region are at present mostly stabilized by grasses and shrubs (Reheis et al., 2005), however, droughts may destabilize large areas leading to increased dust availability (Hiza, 2002). Reheis et al. (2005) noted that changes in local climate and regional conditions lead to variations in the production and delivery of aeolian dust, as evidenced through the stratigraphical record in the Canyonlands area.

The Alamosa Valley PSA sample (N 37°28.453′, W 108° 38.451′, elevation: 2324 m.a.s.l.) was located in a broad, flat valley west of the San Juan National Forest between two mountain ranges in an active dust storm area. Here, wide arrays of sedimentary, plutonic, and volcanic rocks are exposed. The sampling site is located within the San Juan Basin, a rectangular region encompassing northwestern NM, southwest CO, and portions of AZ and UT. The basin itself consists of sedimentary rocks ranging in age from Cambrian to Tertiary, with a semiarid continental climate and saltbrush, sagebrush, and grass vegetation (Stute et al., 1995).

The Little Colorado Corridor (LCC) PSA (N 36° 05.629′, W 111°24.828′, elevation: 1437 m.a.s.l.) is located within the larger Colorado River basin, in an arid to semi-arid region in north central AZ. The sample was taken close to a primitive road and consisted of dark brown dust with a crust. In the Colorado River corridor, exposed fluvial sediment is moved by wind to form sand dunes, and the largest aeolian dune fields occur adjacent to the widest river corridors (Draut, 2012).

The sample from the Chilchinbito Region (N 36°35.017′, W 110°03.671′, elevation: 1700 m.a.s.l.) is located in northeastern AZ on the border between Province 2 and 3 as defined in Bennett and DePaolo (1987). The Chilchinbito region is in the north-central portion of the Navajo Indian Reservation and is comprised of sedimentary rocks ranging from Triassic, Jurassic, and Cretaceous in age (Beaumont and Dixon, 1965). Flat-lying rocks within the Chilchinbito quadrangle comprise a southeastward oriented plateau, which is forested with piñon pine, juniper and ponderosa pine (Beaumont and Dixon, 1965). High mesas intersected by canyons are located throughout the Chilchinbito region; these canyons are bare, whereas the mesas are moderately vegetated. Two-thirds of the land area in northeast AZ had been significantly disturbed due to heavy livestock grazing by the early 1930s (Grahame and Sisk, 2002; Neff et al., 2008). The effects of heavy livestock grazing have been noted to result in a loss of soil stability and a subsequent increase in aeolian erosion of soil (Belnap and Gillette, 1998; Neff et al., 2005).

Finally, two samples were collected from eastern NV: Cadiz Lake, a dry playa consisting of very fine sediment (N 37°51.506′, W 115°15.198′, elevation: 1508 m.a.s.l.) and Coyote Springs, an aeolian dune (N 36°59.320′, W 114°58.700′, elevation: 765 m.a.s.l.). Cadiz Lake, NV is comprised primarily of clay minerals in a continental saline environment (Droste, 1961), whereas Coyote Springs, NV is alluvium underlain by limestone (Reheis et al., 2009).

### 2.4. Mojavia

The Mojave Desert region (Mojavia) consists of mountain ranges containing a broad range of rock types that are bordered by alluvial fans and valley floors that have dry riverbeds and lakebeds serving as sources of dust (Goldstein et al., 2008). Eleven samples were collected for this study from the Mojavia region, ranging in elevation from 272 to 2712 m.a.s.l., with dust originating from alluvial and/or playa deposits. Newly deposited alluvial and playa sediments have been attributed as the primary local sources of dust in the arid southwest (Mojavia) region (Reheis and Kihl, 1995), with previous research indicating that dust from sparsely vegetated alluvial plains and fans are larger sources of modern dust than playas (Reheis and Kihl, 1995), excluding the artificially drained Owens Lake (Reheis, 1997). Mojavia is characterized by a variety of geologic surfaces, including assorted rock types, playas, sand dunes, alluvial fans and riverbeds (Reynolds et al., 2006b). Dust from this region originates from diverse lithology, including carbonate (i.e. limestone and dolomite), volcanic (i.e. rhyolite and basalt), and metasedimentary rocks (i.e. schist and calcareous rocks) (Reheis et al., 2002a). Previous geochemical characterization of dust from the

Mojave Desert showed that dust recently deposited in the region is chemically and mineralogically similar over a large area and different geologic substrates (Reynolds et al., 2006b), suggesting that the deposited dust is originating from a well-mixed source, either in the atmosphere or in fluvial, alluvial and lacustrine environments before dust uplift.

#### 2.4.1. McCullough Range, NV

The McCullough Range PSA sample (N 35°30.709′, W 115°04.399′, elevation: 1314 m.a.s.l.) was collected in close proximity to a dry riverbed near the McCullough Mountains in southern NV. The McCullough Range is composed of Early Proterozoic gneiss derived from metasedimentary and meta-igneous protoliths (De Witt et al., 1989). Previous field exploration of the McCullough Range found fibrous blue and green amphibole minerals in fractures inside Miocene granitoid plutons, which have been noted to be a source of dust transported from large alluvial fans toward the city of Henderson, NV (Buck et al., 2013). The fibrous amphiboles found in southern NV and transported by aeolian processes and increased anthropogenic activity (i.e. driving off-road) may increase human exposure to known human carcinogens (Buck et al., 2013).

#### 2.4.2. Bare Mountain, Crater Flat, Goldfield & Tonopah, NV

The samples from Bare Mountain (N 38°08.689′, W 116° 37.162′, elevation: 1724 m.a.s.l.) and Crater Flat (N 36° 48.797′, W 116°43.609′, elevation: 900 m.a.s.l.) were taken from alluvium underlain by limestone and metamorphic rocks, respectively, while samples from Goldfield (N 37° 41.769′, W 117°14.203′, elevation: 1781 m.a.s.l.) and Tonopah (N 38°04.321′, W 117°07.252′, elevation: 1675 m.a.s.l.), NV were collected from a dry playa and alluvium, respectively. For Goldfield, the local substrate is basalt, while the Tonopah area is underlain by rhyolite and granite. All areas were previously examined for trace elemental composition in Reheis et al. (2009), who concluded that the composition of these dust source areas are influenced by anthropogenic activity and emissions from the now desiccated Owens Lake in 1926. The change in geochemical composition of modern dust from these locations provides evidence of anthropogenic influence upon the local dust cycle. Lastly, the sample collected from Trail Canyon, NV (N 37°54.114′, W 118°08.451′, elevation: 1574 m.a.s.l.) was located on a dry wash with a cemented crust (~3 cm), similar to young Holocene deposits with weakly developed desert pavements described in Reheis et al. (2009).

#### 2.4.3. Stooge range and white mountains

The Stooge Range, CA PSA sample (N 37°00.086′, W 118°12.909′, elevation: 1177 m.a.s.l.) and the White Mountains, CA PSA sample (N 37°21.576′, W 118°10.581′, elevation: 2712 m.a.s.l.) were collected from the Mono Basin, which lies adjacent to the eastern side of the central Sierra Nevada, and is the highest hydrographically closed basin in the Basin and Range province (Reheis et al., 2002b). The Stooge Range sample was collected from an alluvial fan in the Owens Valley, CA, and the White Mountains sample was collected from a high elevation bristle cone pine forest. The granitic and metamorphic rocks composing the Sierra Nevada underlie the Mono Basin, and these rocks are overlain by Miocene to Holocene volcanic rocks and interbedded alluvial and lacustrine sediments (Gilbert et al., 1968). Previous studies have established that the Owens Valley dust has anomalous metal enrichments (Reheis et al., 2009), and dust from this region is transported regionally (Reheis et al., 2002b) as a result of the desiccation of Owens Lake during the 20th century (Gill, 1996; Saint Amand et al., 1986).

#### 2.4.4. Turtle Mountains, Kelso Dunes, Upper Silver Lake, CA

Three Mojavia samples were collected from: Turtle Mountains (N 34°12.511′, W 114°37.059′, elevation: 308 m.a.s.l.), Kelso Dunes (N 34°53.689′, W 115°42.150′, elevation: 795 m.a.s.l.), and Upper Silver Lake, CA (N 35°18.353′, W 116°05.212′, elevation: 271 m.a.s.l.). This portion of the Mojave is characterized by its diverse geology and setting: playas, paleolake deposits, volcanic deposits, alluvial fans, and riverbeds (Reynolds et al., 2006a). The Turtle Mountains site was located in an alluvial fan surrounded by dry shrubs and pumice. Here, the sampled dust was a mixture of fine and coarse-grained material that had been covered by a slight crust. The Kelso Dunes sample was collected from a large sand dune in the center of a valley, and consisted of fine-grained, homogeneous dust that appeared to have been thoroughly reworked by aeolian processes. The Upper Silver Lake site is located atop granitic intrusions and metamorphic rocks, adjacent to the Cima Volcanic field.

### 2.5. San Luis Valley

The San Luis Valley (SLV) is a high elevation (~2300 m. a.s.l.) desert valley that extends from south-central CO into NM. High concentrations of mafic rock material are present throughout the SLV area due to mid-Tertiary (Brice, 2013) and Cenozoic volcanic activity (Sims et al., 2007). The climate in the SLV is influenced by the Rocky Mountain winter-dominant precipitation regime and the dry summer-dominant precipitation regime of the southwestern deserts and Great Plains in North America, and receives less than 178 mm/yr primarily during the summer monsoon (Brice, 2013). Erosion rates in western NM have been estimated to range from 1.7 to 5 mm/k.y. using U-series isotopes and He cosmogenic ages (Sims et al., 2007).

The Twin Lakes (N 35°42.080′, W 108°46.465′, elevation: 1954 m.a.s.l.) and Newcomb (N 36°16.762′, W 108° 42.281′, elevation: 1695 m.a.s.l.) samples were collected from roadside locations, both from dark, relatively homogenous dust. Aeolian transported dust from the SLV has been examined in previous studies (Janke, 2002; Madole et al., 2008). An area known as the "sump" in the southwest region of the Rio Grande basin has been identified as a major source of aeolian sediment transported to the Great Sand Dunes National Park and Preserve (GSDNPP) in southern CO (Madole et al., 2008). The pri-

mary controlling factor in dust transport to GSDNPP is changes to the hydrologic cycle (i.e. flooding and variations in the water table) (Madole et al., 2008).

## 3. METHODS

Consistent with the methods described in Gaiero et al. (2003), the upper 5 cm of surface topsoil was sampled with a trowel and stored in sterile Whirl-Pak sampling bags. While surface sediments collected in this way are a good proxy for potential source material transported atmospherically, the samples collected here are not strictly representative of soils derived exclusively from local parent materials due to the likelihood of some input from exogenic sources. The samples collected here are representative of sediment available for transport and were previously undocumented from a geochemical perspective. Surface material was collected from one location at each site from relatively open areas with little overstory vegetation, and evidence of aeolian reworking through ripple marks or dune migration. For each location, relatively large quantities of material (at least 500 g) were collected to ensure that once sieved and passed through the filtration unit, a sufficient amount of fine-grained material for the analysis of both isotopic and elemental compositions would remain. Isotope compositions of sediments can vary based on size fraction, most notably with Sr isotopes (Dasch, 1969). The Hf isotope composition of dust transported through the atmosphere also varies based on distance from the source (Aarons et al., 2013). To account for these potential variations, we separated each sample into three different size fractions (0.2–10 um, 10–30 um, 30–63 um) to differentiate between regional (larger particles) and global (smaller particles) transportable dust. The sampling campaign was carried out during the summer of 2014, with laboratory analysis accomplished in the subsequent months.

First, bulk samples were passed through a sequence of Fisherbrand U.S. Standard Stainless Steel Test Sieves, the coarsest of which removed all woody debris and whole rock materials (i.e. pebbles, cobbles) that could not be removed in the field. The finest mesh selected all particles <63 μm in diameter, which were rinsed from the collection pan with Super Q $H_2O$ (>18.2 MΩ cm) and transferred in centrifuge tubes to ISO class 7 clean room at the University of Michigan's Department of Earth and Environmental Sciences. All analyses unless otherwise noted were conducted at the University of Michigan. All further sample preparation and subsequent column chemistry was conducted within ISO class 4 laminar flow hoods.

Each sample was separated into three size fractions: fine: 0.2–10 μm; medium: 10–30 μm; and coarse: 30–63 μm using pre-cleaned, Savillex hydrophobic PTFE filters. After filtration, samples were rinsed and weighed into 3 mL Teflon beakers. Sample dissolutions were carried out using an Ethos Milestone Microwave Labstation, which holds up to ten 100 mL Teflon sample vessels in a segmented rotor assembly. For our purposes, two sample solutions at a time were placed inside each vessel, sealed tightly within 3 mL Savillex beakers. Prior to dissolution, samples had been pre-treated with two dry-downs in 1 mL 16 M $HNO_3$ +1 mL 25 M HF + 0.5 mL Optima-grade $H_2O_2$ at ~110° C (all acids were double-distilled (Fisher Optima or Seastar)); however, some of the garnet- and zircon-bearing samples remained cloudy or yielded precipitates after the second dry-down, and therefore required up to 4 dry-downs to completely eliminate fluorides.

During the microwave session, power was continually increased or decreased as dictated by conditions in a reference vessel containing a temperature-monitoring probe. Sealed within the reference vessel were two beakers and an acid mixture identical to those present in other vessels. We employed a three-step dissolution process; for the first step, samples were dissolved in an acid mixture of 1 mL 16 M $HNO_3$ + 1 mL 25 M HF, with the second solution containing 2 mL 25 M HF, and the third 2 mL 6 M HCl. For each step, the microwave heating regime consisted of a 10 min warm-up to 200°C, 30 min at 200°C, and a 20 min cool-down to room temperature. These solutions were dried down and then re-acidified in 1 mL of 9 M hydrochloric acid in preparation for chemical separation via ion-exchange chromatography, wherein the techniques of Aciego et al. (2009) were used to isolate the elements Sr, Nd, and Hf for isotopic analyses. A 10% split from the total digestion of sediment reserved for analysis of major and trace element concentrations.

Sr and Nd isotopic compositions were measured on a Thermo Scientific Triton PLUS Thermal Ionization Mass Spectrometer (TIMS) following the same procedures employed by Blakowski et al. (2016), Stevenson et al. (2016), Clinger et al. (2016) and Aarons et al. (2016b). Sr measurements were normalized to $^{86}Sr/^{88}Sr = 0.1194$ to correct for mass fractionation. The long term Sr isotopic standard NBS-SRM-987 measured $^{87}Sr/^{86}Sr = 0.710245$ ($2\sigma = \pm 2.3 \times 10^{-5}$, n = 208), and the BCR-2 Unites States Geological Survey (USGS) basalt rock standard was $^{87}Sr/^{86}Sr = 0.705012$ ($2\sigma = \pm 6.1 \times 10^{-5}$, n = 2), in agreement with other reported literature values (e.g., $^{87}Sr/^{86}Sr = 0.705000$, Jweda et al., 2015). Nd isotopic ratios were also corrected for mass fractionation using $^{146}Nd/^{144}Nd = 0.7129$, with mass 149 monitored for potential Sm interference. The long term Nd isotopic standard JNdi-1 (10 ng) measured $^{143}Nd/^{144}Nd = 0.512101$ ($2\sigma = \pm 2.4 \times 10^{-5}$, n = 19), and the USGS rock standard BCR-2 had a $^{143}Nd/^{144}Nd$ ratio of 0.512641 ($2\sigma = \pm 3.3 \times 10^{-6}$, n = 2), also in agreement with previously established measured ratios (e.g., $^{143}Nd/^{144}Nd = 0.512637$, Jweda et al., 2015). For both Sr and Nd, the measured blanks were considered to be negligible. Below, the Sr compositions of PSA materials continue to be reported in $^{87}Sr/^{86}Sr$, while Nd ratios are reported in terms of their deviations from the Chondritic Uniform Reservoir (CHUR) evolution line, wherein $\varepsilon_{Nd} = (((^{143}Nd/^{144}Nd)_{measured}/(^{143}Nd/^{144}Nd)_{CHUR}) - 1 \times 10^4$, using the present-day CHUR value of 0.512638 (DePaolo and Wasserburg, 1976).

Hafnium (Hf) isotope composition measurements were performed at the University of Wyoming's High-Precision Isotope Laboratory on a Thermo Scientific Neptune Plus Multiple Collector Inductively Coupled Plasma Mass Spectrometer (MC-ICP-MS) as detailed in Arendt et al. (2014) and Blakowski et al. (2016). During this analytical session,

amplifier gains were evaluated at the start of each day, and baselines were measured prior to each sample. Instrumental mass fractionation was corrected using $^{179}Hf/^{177}Hf = 0.7325$ and an exponential fractionation law, and masses 182, 175, and 172 were monitored for W, Lu and Yb interferences. The corrections to $^{176}Hf/^{177}Hf$ were less than 100 ppm. All isotopic measurements were further normalized externally by bracketing each sample with the Hf standard reference material JMC-475 (accepted value of $^{176}Hf/^{177}Hf = 0.28216$) (Vervoort and Blichert-Toft, 1999). The USGS BCR-2 rock standard measured $^{176}Hf/^{177}Hf = 0.282894$ ($2\sigma = 5.4 \pm 10^{-6}$, n = 2), in agreement with previously reported values (e.g., $^{176}Hf/^{177}Hf = 0.282869$, Jweda et al., 2015). Hereafter, Hf isotope ratios are also presented in epsilon notation, with $\varepsilon_{Hf} = ((^{176}Hf/^{177}Hf)_{measured}/(^{176}Hf/^{177}Hf)_{CHUR}) - 1 \times 10^4$, with CHUR = 0.282772 (Blichert-Toft and Albarède, 1997). We note that certain samples did not contain sufficient Hf concentrations to yield precise isotope measurements, which reduced our Hf dataset compared to the number of Sr and Nd data points.

Analyses of major and trace element concentrations were carried out on the Thermo Scientific ELEMENT2 ICP-MS at the Keck Laboratory operating in pulse counting mode. Here, concentrations were calculated using a three-point calibration curve, described in further detail by Aciego et al. (2015). Every five samples, an acid blank and a standard of known concentration (NIST1640a) were run as unknowns. All standards reproduced within error of the calibration curve, and the acid blank was below detection limits. For the smallest elemental concentrations (e.g. Fe), the total procedural blank could account for approximately 10% of the concentration measured (see Table S1). Trace element concentrations reported in Fig. 2 were normalized to the average continental crust concentration (Wedepohl, 1995). For example, the reported concentration for zinc (Zn) is:

$$Zn_{reported} = [Zn]_{sample}/[Zn]_{crust}$$

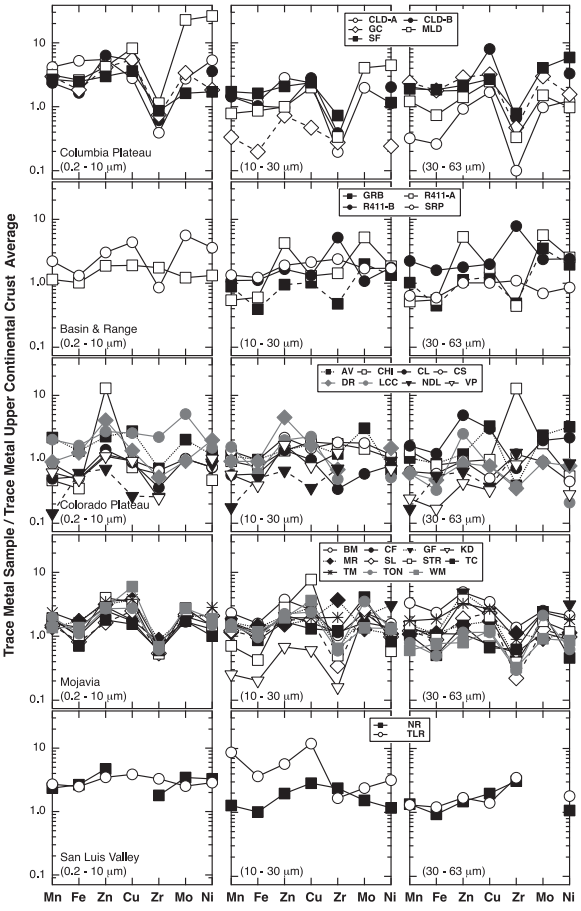


Fig. 2. Trace metal concentrations measured in PSA samples normalized to the average composition of the upper continental crust for each of the three grain size fractions analyzed in this study (with grain size increasing from left to right). Samples are separated by geologic province defined in Fig. 1. See Table 1 for sample codes, and supplementary Table S1 for measured trace element concentrations.

## 4. RESULTS

### 4.1. Trace metal concentrations

The trace metal concentrations for the dust samples measured are similar to the average continental crustal concentrations reported in Taylor and McLennan (1985) and Wedepohl (1995). The fine (0.2–10 μm) size fraction samples from the Columbia Plateau are in general more enriched in all trace metals compared to the remaining provinces. Zirconium (Zr) concentrations appear to be progressively more depleted with increasing particle diameter in the sediment samples from the Columbia Plateau, which was unexpected given that the high density mineral zircon is chemically resistant to weathering, which tends to result in coarser, sandy sediments containing a greater abundance of zircons (Aarons et al., 2013). Also identifiable for the Columbia Plateau province is the higher enrichment of trace metals in the fine fraction (0.2–10 μm) versus the two larger grain size fractions (10–30 and 30–63 μm, respectively). Likewise, the trace metal patterns for the finest grain size fraction from the Mojavia province are more homogeneous compared to the two larger grain size fractions (Fig. 2). The variability of trace metal concentrations for the Dubois, WY sample (R411) and the SRP sample (SRP-060514) in the Basin & Range are similar for the two larger grain size fractions, whereas the pattern is much different for the smallest grain size fraction (Fig. 2), possibly due to degree of weathering. The trace element concentration patterns for samples from the Colorado Plateau are fairly similar, with small excursions for a particular element (Zn in the fine fraction and Zr in the coarsest fraction from Chilchinbito Region, AZ (CHI-061114)). For the San Luis Valley, samples from the Newcomb Region, NM (NR-061114) are fairly homogeneous with respect to trace element concentration patterns, whereas samples from the Twin Lakes Region, NM (TLR-061114) show higher variability across the different size fractions measured here (Fig. 2).

### 4.2. Phosphorus nutrient supply

The importance of phosphorus (P) inputs from atmospherically transported dust for nutrient dynamics and soil productivity varies across ecosystems; some soils are known to be highly reliant upon P (and other critical nutrients such as Ca, Mg, K) delivered by aeolian dusts (Aciego et al., 2017; Chadwick et al., 1999; Porder et al., 2007; Yu et al., 2015), while others acquire and sustain sufficient P from the weathering of local parent material (Hudson-Edwards et al., 2014; Okin et al., 2004; Pett-Ridge, 2009). In terms of particle size, it has been suggested that potentially limiting rock-derived nutrients such as P are primarily associated with the finest fraction (silt- and clay-sized) soil particles in the arid and semiarid regions of the western US (Neff et al., 2005; Pett-Ridge, 2009).

Indeed, the majority of finer grained materials (0.2–10 μm and 10–30 μm) analyzed in this study appear to be slightly more enriched in P with respect to the average composition of the upper continental crust, compared to the larger (30–63 μm) grain size fraction (Fig. 3). Although the samples analyzed in this study as a whole are not significantly enriched in P, the calculated mean $P_{sample}/P_{UCC\ avg}$ values ranges between 1 and 2 for each of the grain size fractions (0–3 considering calculated standard deviations). Thus, it is possible that these natural soil dusts provide a greater contribution of P than previously thought to nutrient-poor ecosystems downwind.

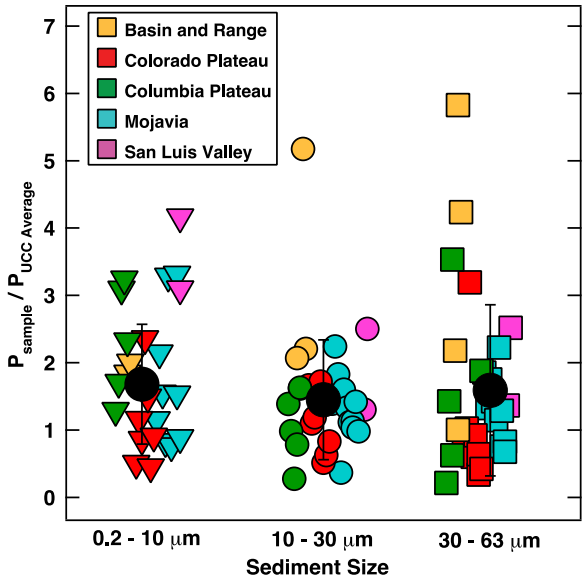


Fig. 3. Phosphorus concentrations measured in each of the three grain size fractions normalized to the average composition of the upper continental crust. Large black circles define the calculated mean value (±1 S.D.) for each grain size fraction. We note that the 10–30 μm and 30–63 μm grain size fractions for the R411 samples (particularly for R411-A), which are composed of young, unconsolidated moraine material from glacial retreat in the Wind River Range (WRR), are abnormally enriched in P (which is also the case for Zn in R411-A and Zr in R411-B). Otherwise, we observe slightly higher phosphorus concentrations in the smallest grain size fraction, with the majority of 30–63 μm samples either depleted in phosphorus or concentrated around $P_{sample}/P_{UCC\ avg} = 1$. Note, the data are slightly offset on the x-axis by locations so that all the data are visible; otherwise, there is no variation other than difference in size fraction. See supplementary Table S1 for measured P concentrations.

### 4.3. Sr-Nd isotopes

Tracing the sources and pathways of dust is possible using radiogenic isotope compositions (Sr, Nd) of the dust preserved in the paleoclimate record (Basile et al., 1997; Delmonte et al., 2010; Grousset and Biscaye, 2005; Revel-Rolland et al., 2006). Past work by Biscaye et al. (1997), Kanayama et al. (2002), and Chen et al. (2007) has provided PSA isotopic compositions in the Northern Hemisphere from both major and minor sources. Previous work in the western US include Nd isotope data from a wide range of rock material from different geologic provinces (i.e. Bennett and DePaolo, 1987) and Sr isotope data from a southwest desert transect (Goldstein et al., 2008). Sr-Nd isotope compositions of bulk rock from the American West is available in the GEOROC database (GEOROC database: http://georoc.mpch-mainz.gwdg.de), however the materials measured in this study represent older, weathered sediment readily available for transport rather than bulk rock composition or young volcanics. Our PSA data shows a clear mantle-derived end-member from the Columbia Plateau (CP-NW) (see Fig. 4) which spans a similar range in Sr-Nd isotope composition compared to the whole rock dataset from GEOROC (individual references can be found in Supplementary Table S2 and are included here in the reference list). Our compositions however, are more tightly clustered compared to the GEOROC isotope compositions allowing for more precise tracing of dust sources (Fig. 4).

The Sr and Nd isotope compositions for each region range from:

(i) Columbia Plateau: $^{87}Sr/^{86}Sr = 0.703699$ to 0.715699 and $\varepsilon_{Nd} = -7.3$ to 2.4.
(ii) Basin & Range: $^{87}Sr/^{86}Sr = 0.710365$ to 0.716247 and $\varepsilon_{Nd} = -13.0$ to −9.4.
(iii) Mojavia: $^{87}Sr/^{86}Sr = 0.707583$ to 0.717167 and $\varepsilon_{Nd} = -15.5$ to 1.3.
(iv) Colorado Plateau: $^{87}Sr/^{86}Sr = 0.707987$ to 0.717320 and $\varepsilon_{Nd} = -14.5$ to −8.7.
(v) SLV: $^{87}Sr/^{86}Sr = 0.712177$ to 0.740236 and $\varepsilon_{Nd} = -26.6$ to −8.5.

The most radiogenic sample with respect to Nd isotope composition is from Christmas Lake Valley dunes (sample B, $\varepsilon_{Nd} = 2.4$), whereas the Sr isotope composition of the Christmas Lake Valley site (sample A & B) is the least radiogenic. One sample from the SLV (Twin Lakes Region, NM) has a very unradiogenic Nd isotope ratio ($\varepsilon_{Nd} = -26.6$, $2\sigma = 0.2$). The samples from the Columbia Plateau (CP-NW) have predominately volcanic Sr-Nd isotope signatures.

When compared to the Sr-Nd isotope compositions of rock samples from the GEOROC database, there is evi-

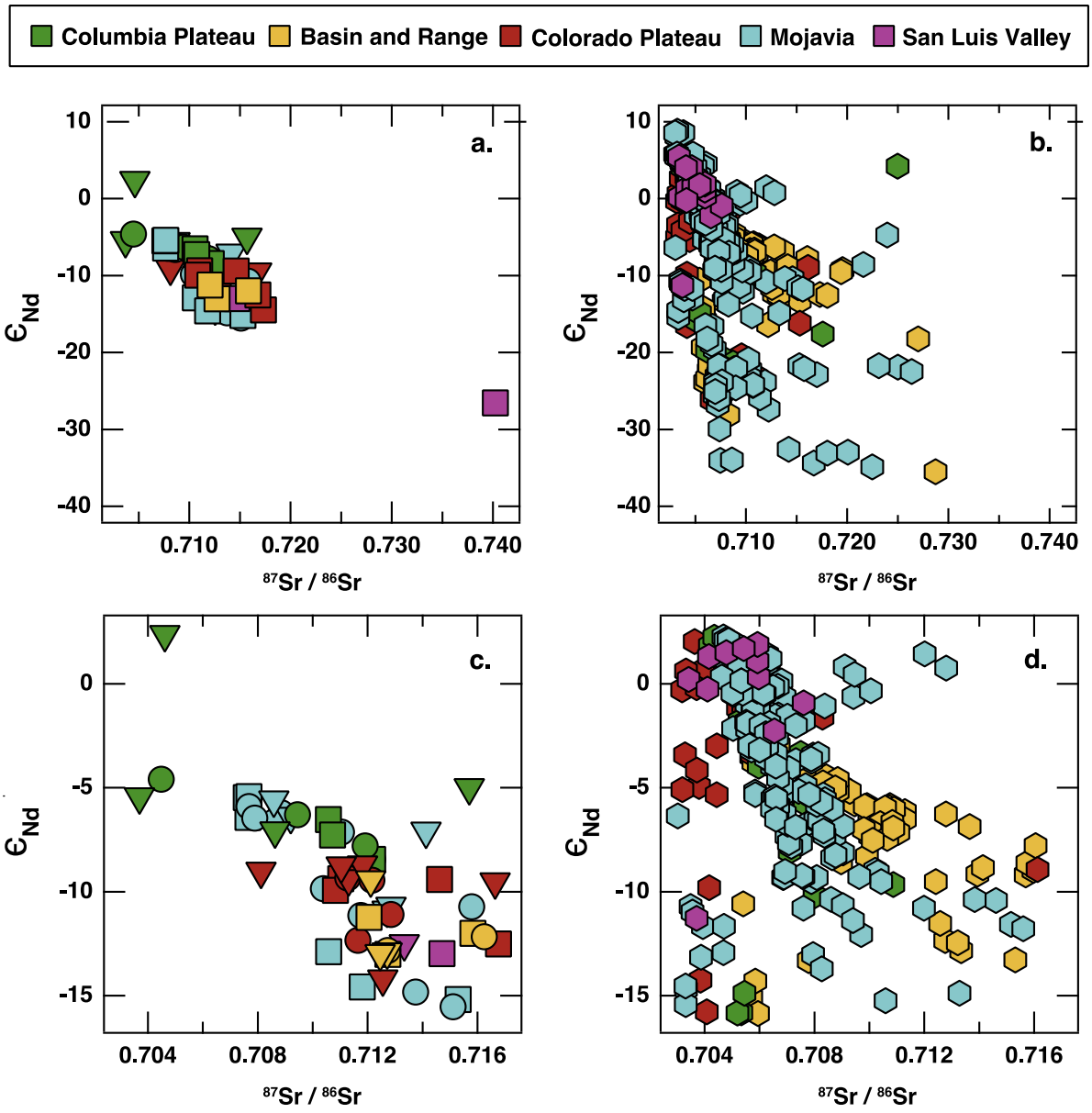


Fig. 4. Sr-Nd isotopic compositions of the PSA samples compared to whole rock isotopic values obtained from the GEOROC database; (a) $^{87}Sr/^{86}Sr$ versus $\varepsilon_{Nd}$ compositions of PSA samples from this study, with the three size fractions differentiated as follows: 0.2–10 μm by triangles; 10–30 μm by circles; 30–63 μm by squares; (b) $^{87}Sr/^{86}Sr$ versus $\varepsilon_{Nd}$ compositions of whole rock samples from previously published geochemical studies in the western U.S.; (c) enlarged version of plot 4a, having removed TLR0611-30 (which is highly radiogenic w.r.t. Sr) to better showcase the isotopic variations in our dataset; (d) portion of the whole rock data using identical isotopic ranges from the enlarged plot 4c from the GEOROC database sources (and references therein). See supplementary Tables S1 and S2 for measured radiogenic isotope compositions of samples measured in this study and samples from the GEOROC database respectively.

dence of intensive physical/chemical weathering (via the Sr isotope composition) in PSA samples originating from the Colorado Plateau region (Fig. 4), whereas the Nd isotope compositions span a similar range to the GEOROC set. We do observe similar isotopic signature ranges for our PSA data compared to the GEOROC data from the Basin & Range and Mojavia (Fig. 4).

### 4.4. Nd-Hf isotopes

The Nd and Hf isotope compositions of environmental samples can be used to indicate the degree of continental weathering (Bayon et al., 2006; Rickli et al., 2010; Rickli et al., 2009; van de Flierdt et al., 2007) or the transport distance of sediment and dust (Aarons et al., 2013). The

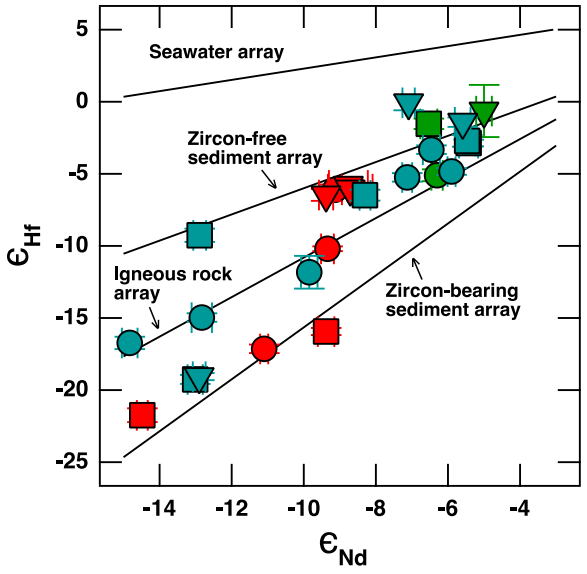


Fig. 5. $\varepsilon_{Nd}$ versus $\varepsilon_{Hf}$ compositions of the PSA samples analyzed in this study, with the various provinces following the same color scheme described in the legend of Fig. 4. Again, the three size fractions are represented as follows: 0.2–10 μm by triangles; 10–30 μm by circles; 30–63 μm by squares. Plotted lines for the zircon-bearing sediment ($\varepsilon_{Hf} = 1.80$, $\varepsilon_{Nd} = 2.35$), zircon-free sediment ($\varepsilon_{Hf} = 0.91$, $\varepsilon_{Nd} = 3.1$), igneous rock ($\varepsilon_{Hf} = 1.37$, $\varepsilon_{Nd} = 2.89$), and seawater ($\varepsilon_{Hf} = 0.39$, $\varepsilon_{Nd} = 6.2$) arrays as defined by Bayon et al. (2009). See supplementary Table S1 for measured radiogenic isotope compositions.

observed Hf and Nd isotopic arrays of earth's oceans, sediments and igneous rocks (Fig. 5) are different due to incongruent weathering of continental crust (Piotrowski et al., 2000; van de Flierdt et al., 2002; van de Flierdt et al., 2007). The mineral zircon's resistance to weathering results in a low $^{176}Hf/^{177}Hf$ ratio imparted to the more zircon-rich portions of the sedimentary system (Vervoort et al., 1999), and the Hf isotope composition can therefore be used to indicate the proximity of the sediment or dust from source (Aarons et al., 2013).

The ranges in Hf isotope compositions of PSA's for dust measured from each region measured are below (we have no Hf data for the SLV region):

(i) Columbia Plateau: $\varepsilon_{Hf} = -5.1$ to $-0.6$.
(ii) Basin and Range: $\varepsilon_{Hf} = -19.3$ to $-15.0$.
(iii) Mojavia: $\varepsilon_{Hf} = -16.7$ to $-0.1$.
(iv) Colorado Plateau: $\varepsilon_{Hf} = -21.7$ to $-5.8$.

Samples originating from the Colorado Plateau region exhibit the strongest weathering signal, with the fine fraction samples plotting along the zircon-free array as expected (Fig. 5). Meanwhile, the medium (10–30 μm) and coarse (30–63 μm) samples from the same region are significantly less radiogenic, plotting along the igneous rock and zircon-bearing sediment arrays. There are no obvious discernible trends in Nd-Hf isotope composition of PSAs from the Columbia Plateau, all samples plot close to the zircon-free array (Fig. 5). Samples originating from Mojavia span the largest range in Nd-Hf isotope composition; one fine (0.2–10 μm) sample plots between the zircon-free sediment array and the seawater array, and the medium and coarse fraction are between the zircon-bearing sediment array and the zircon-free sediment array.

## 5. DISCUSSION

A number of studies indicate that dust is an important contributor to regolith composition, both in arid and semi-arid climates of the western US (Aciego et al., 2017; Blank et al., 1999; Lawrence and Neff, 2009; Lawrence et al., 2011; Reynolds et al., 2006c). As many of these studies have noted, incorporation of dust into regolith has consequences for geomorphic and geochemical studies, and tracing the sources and pathways is important for studies spanning from the disciplines of biogeochemistry, geobiology, and ecology.

### 5.1. Importance of trace metal input to ecosystems

Soils are the main sink and source of trace metals, and it has been established that the transition from bedrock to soil is the largest source of nutrients and trace metal input (As, Cd, Cr, Cu, Ni, Pb, and Zn) (Ferrier et al., 2011). However, it has also been suggested that aeolian dust input is an increasingly important source of critical nutrients, in both ecosystems that are depleted due to chemical and physical weathering (Chadwick et al., 1999; Porder et al., 2007; Yu et al., 2015), and in actively eroding mountain systems (Aciego et al., 2017). In the sedimentary system, clay-sized particles are generally known to absorb higher metal concentrations than any other soil fraction (Farrah and Pickering, 1977; Gupta and Chen, 1975); therefore, we expect the highest trace metal concentrations to be found in the smallest size fraction materials.

Elements considered to be immobile are present in small quantities in most regolith, and the total concentration of immobile elements in regolith can be altered by even very small contributions of atmospherically transported dust, especially if the dust is rich in these elements (Aciego et al., 2017). Quantifying the influence of dust on regolith composition (and hence on chemical and physical erosion rates inferred from regolith composition) thus requires estimates of dust composition and rates of dust incorporation in regolith. The trace element compositions from each source region measured here provide a preliminary survey of trace metal enrichment in transportable dust in the American West, with select regions in our dataset being particularly distinguishable in terms of elemental composition (i.e. Mojavia, Columbia Plateau, Fig. 2). Further comparison of the trace element compositions to the underlying bedrock would provide more information about the fractionation of heavy trace metals that occurs during chemical and physical weathering, which would have implications for trace metal sources, sinks, fluxes, and uptake to ecosystems.

### 5.2. Phosphorus concentrations: implications for biogeochemical cycles

The role of mineral dust as a source of P varies across regions, however, the transport of dust is particularly important in ecosystems underlain by highly weathered and/or nutrient-depleted parent materials (Hudson-Edwards et al., 2014; Okin et al., 2004; Pett-Ridge, 2009). Phosphorus is often a limiting nutrient in terrestrial environments (Okin et al., 2004), and recent work suggests that natural eroding processes in mountain landscapes result in a depletion of P, which instead is delivered via dust deposition from near and far sources (Aciego et al., 2017). For example, P-enriched aeolian dusts deposited in old and weathered, geomorphically-stable landscapes in Hawaii and the Amazon basin significantly subsidizes ecosystem nutrient budgets (Chadwick et al., 1999; Okin et al., 2004). Likewise, P derived from atmospherically transported dust input may be important in young ecosystems developed on active geomorphic settings with P-deficient parent materials (Okin et al., 2004). Human-related disruption to the P cycle as a result of the use of fertilizers in the second half of the twentieth century (Mikkelsen and Bruulsema, 2005) may also lead to increasing P concentrations in sediments available for aeolian transport (Neff et al., 2008). Ecosystems already rich in P may receive additional input from the deposition of P-rich dust, and this may lead to eutrophication in lakes as observed in the Sierra Nevada (Homyack et al., 2014) or increased biological activity as observed in lake ecosystems in the intermountain West (Brahney et al., 2015). The implications of increased P-input to mountain streams, lakes, and terrestrial ecosystems are not fully understood, and as such, this topic is worthy of further investigation, as fertilization of these environments is capable of contributing to $CO_2$ sequestration and therefore the carbon cycle (Hudson-Edwards et al., 2014).

Quantifying P variability in dust sources from the American West allows extrapolation of nutrient fluxes from exogenic sources. Aciego et al. (2017) used existing measurements of soil production rates in an ecosystem with bulk geochemistry of bedrock to estimate nutrient flux from bedrock, and dust flux rates combined with geochemical measurements to estimate nutrient flux from aeolian dust. Previous work has established that agricultural activity combined with either natural or human-induced changes in climate resulted in the increased dust input to a North American glacier (Aarons et al., 2016a). Providing constraints upon the P concentrations in PSAs of dust to marine and terrestrial biospheres can quantify P delivered to ecosystems as a result of higher anthropogenic activity and amplified drought. Higher dust availability has implications for biogeochemical cycles, and although future dust sources and transport pathways are difficult to predict, it is likely that ecosystems will be influenced by increased P availability.

### 5.3. Compositional variability: applications of radiogenic isotopes

Over the last two decades, Sr and Nd isotopes have been used to determine the geographic provenance of aeolian dust deposited in natural sinks such as dry lake beds, sediment cores, and snow and ice layers (Basile et al., 1997; Biscaye et al., 1997; Delmonte et al., 2004; Grousset and Biscaye, 2005; Grousset et al., 1992). While it is not always possible to pinpoint the exact origin of a mineral dust sample, the unique physical and isotopic properties of materials featured in this study have allowed us to narrow down and characterize a number of potential dust sources in the American West.

The American West consists of a wide variety of rock types and ages, and the resulting $^{87}Sr/^{86}Sr$ and $\varepsilon_{Nd}$ composition span a broad range. While our PSA samples resemble areas of the existing whole-rock geochemical data from the GEOROC database, they provide a more highly refined Sr-Nd catalog for determining provenance of dust in modern and paleoenvironmental archives. Comparing the Sr-Nd isotope composition of aeolian dust to whole-rock data will not provide accurate characterization, as the incongruent weathering of rubidium (Rb) and Sr leads to higher $^{87}Sr/^{86}Sr$ ratios in more weathered samples (Dasch, 1969). The Nd isotope composition should not vary based upon the degree of weathering, as Nd is released congruently throughout the weathering process (Garcon et al., 2013; Goldstein et al., 1984).

In our sample set, the Sr isotope composition of dust from the same geologic province as whole rock samples are more radiogenic, most notably for samples from the Colorado Plateau and the SLV (Fig. 4). We do see more refinement in the Nd isotope composition of the majority of our PSA samples ($-15.5 < \varepsilon_{Nd} < -4.6$, excluding one sample from the Columbia Plateau, Fig. 4c) compared to the whole-rock samples (Fig. 4d), however, the more tightly clustered Nd isotope compositions in the PSA versus whole-rock database likely result from the sampling of more confined areas. We note that one PSA sample (TLR0611) from the SLV province differs significantly from observed Sr-Nd isotope composition of the whole-rock samples (Fig. 4a and b). In this case, it is possible that continental-derived dusts and sediments from other localities were transported and deposited to this site, as aeolian reworking, sorting, and transport occurs over time.

In terms of differences in isotope composition with respect to particle size, we do not see significant evidence of physical or chemical weathering resulting in more radiogenic Sr isotope compositions with decreasing particle size (Fig. 4a and c). For example, fine (0.2–10 μm) dust samples from the Columbia Plateau, Mojavia, the Colorado Plateau, and the Basin & Range span similar Sr-Nd isotope compositions as their coarse (30–63 μm) counterparts. This data suggests that the influence of size-dependent fractionation in isotope composition is most substantial for the transition of rock to transportable dust material, rather than the physical and/or chemical weathering of coarse to fine particles.

The erosion of parent material, generation of dust, and subsequent deposition are nearly constant processes in arid and semi-arid environments. Thus, the role of dust input in soil building and composition can be significant depending on the original geochemical composition of a soil sequence (Muhs and Benedict, 2006; Porder et al., 2007; Simonson,

1995). Despite this observation, many geomorphological studies do not take into account the dust inputs due to the lack of dust compositional and flux data. Recent work exploring this problem has found significant influence of dust input upon trace metal concentrations in soils from the American West (Lawrence et al., 2013), however the sources of dust remain unclear.

Desert dust also forces earlier snowmelt in the Rocky Mountains via enhanced solar radiation adsorption (Painter et al., 2007; Painter et al., 2010; Painter et al., 2012; Clow et al., 2016), and the current levels of dust loading are noticeably higher than prior to the mid-1800s (Neff et al., 2008). Decreased snow albedo caused by higher dust loading shortens the duration of mountain snow cover, which in turn advances the peak runoff (Deems et al., 2013; Painter et al., 2007; Painter et al., 2010; Skiles et al., 2012) and therefore has implications for soil aridity and compounding risks for water resources in arid regions in the US. Climate projections for the southwestern US indicate increased aridity; if so, we expect that aeolian dust production and deposition will also continue to increase. Paleorecords can provide valuable information on long-term changes in dust deposition, however the influence of humans upon the dust cycle can be achieved through modern, in-situ monitoring of dust deposition to passive dust collectors (Aciego et al., 2017) and mountain snowpack (Clow et al., 2016). Determining the sources of dust to these collection sites is difficult when only trace element concentrations are taken into consideration, and may be more easily achieved using Sr, Nd, and Hf isotopes.

Other recent work has utilized Sr and Nd isotopes to study the relative contributions of continental dust to potentially nutrient-limited environments, estimating long-term dust input based upon isotope mass balance equations (Li et al., 2016). As soils develop in humid environments, rock-derived elements are rapidly lost. Highly weathered arid environments and montane ecosystems can also be P limited (Aciego et al., 2017), and require exogenic inputs for critical nutrients. Thus, continuous addition of new materials (in the form of dust) to soil is crucial. Estimating the dust flux in constantly evolving terrains is difficult; however, estimations are possible using local regolith production rates, and the concentrations of immobile elements in the regolith, its parent material, and dust (Ferrier et al., 2011). It is therefore possible to apply the known trace element concentrations of soil profiles and PSAs of dust to calculate the mass fraction of dust present in a given ecosystem.

As such, the new dataset presented here complements studies examining the flux of dust from provinces within the American West to potentially nutrient limited ecosystems. For example a lake sediment core study focusing on the influence of increasing dust deposition upon biogeochemical cycles in the western US utilized Paleoproterozoic basement rocks underlying AZ and NM rather than surficial sediment to identify the likely dust sources (Neff et al., 2008). The Sr and Nd isotopic compositions of PSAs presented in this study suggest several possible sources to the high-elevation lakes in the San Juan Mountains, CO such as Crater Flat, NV, Turtle Mountains, CA, Chilchinbito Region, AZ, and the Snake River Plain, ID. This matching of PSAs to dust deposited in lake sediments highlights the strength of our dataset in identifying likely PSAs of dust in existing and future studies.

### 5.4. Insight into continental weathering from Nd-Hf isotopes

While Sr-Nd isotopes are considered the traditional indicator of dust provenance, contributions from global versus regional sources can be distinguished using the lutetium (Lu)-Hf radiogenic isotope system. The relationship of Lu-Hf and Sm-Nd are closely coupled within Earth's mantle; however, the ratio between the two element systems changes significantly during crystallization, subsequently resulting in varied isotopic compositions in Earth surface materials such as rock, regolith, dust, and seawater (Aarons et al., 2013; Bayon et al., 2006; Blakowski et al., 2016; Carpentier et al., 2009; Rickli et al., 2010; van de Flierdt et al., 2004).

Mineral sorting during physical weathering and atmospheric transport can cause changes in the $\varepsilon_{Hf}$ isotope system. The high concentration of Hf in the mineral zircon ($ZrSiO_4$) combined with the low initial Lu/Hf ratio in the mineral leads to very unradiogenic $^{176}Hf/^{177}Hf$ ratios over time (Patchett et al., 1981). As such, a very low $^{176}Hf/^{177}Hf$ ratio is present in the zircon-rich portion of the sedimentary system (Vervoort and Blichert-Toft, 1999), and a more radiogenic $^{176}Hf/^{177}Hf$ ratio is found in finer sediments (i.e. clays, due to the absence of zircon) (Patchett et al., 1984). Additional fractionation in the Hf isotope system occurs with dissolution; the radiogenic global seawater array (see Fig. 5) is a result of the incongruent weathering of Hf (Piotrowski et al., 2000; van de Flierdt et al., 2002; van de Flierdt et al., 2007). As dust is transported through the atmosphere, density-driven fallout of zircons prior to less dense minerals leads to an increase in the $\varepsilon_{Hf}$ composition with distance from the source. This "zircon effect" is applicable to dust transport studies, and it is possible to use the $\varepsilon_{Hf}$ composition to discern transport distance (Aarons et al., 2013). Modeling the "zircon effect" during dust transport suggests that zircon loss occurs between 0 and 3000 km from the source (Aarons et al., 2013). The Nd and Hf isotope compositions of environmental samples can therefore be powerful indicators of continental weathering (Bayon et al., 2006; Rickli et al., 2010; Rickli et al., 2009; van de Flierdt et al., 2007). The endmember $\varepsilon_{Nd}$ and $\varepsilon_{Hf}$ compositions of transportable dust prior to transport provides the information necessary to calculate the relative contributions, and potentially the transport distance of dust.

## 6. CONCLUSIONS AND IMPLICATIONS

We report preliminary geochemical characterizations of significant dust sources throughout the American West. Future work on the effects of climate change and human activity on dust production and deposition in places ranging from terrestrial biospheres to mountain snowpacks and glaciers will require endmember isotope compositions of transportable dust from known dust source areas. We find that the majority of the finest grained samples analyzed

in this work are slightly more enriched in P compared to coarser grained samples. We hypothesize that information on potentially limited nutrients, such as P present in dust source areas, will prove valuable in future studies focusing on the biogeochemical implications of increased dust availability and transport in the American West. Additionally, the Sr-Nd isotope compositions of PSAs of dust in this study are more constrained compared to GEOROC data of whole rock samples, and provide a more accurate characterization of dust available for transport in specific regions. In general, we do see a shift towards more radiogenic Sr isotope compositions with the degree of weathering between the whole rock and dust data. Although there is considerable overlap in Sr-Nd isotope compositions of dust PSAs in this study, there are distinguishable patterns in the Sr-Nd arrays of the different geologic provinces studied. Hf isotope composition combined with satellite observational data and dust size distribution will provide additional insight into dust provenance and transport distance. Future work will involve expanding the chemical characterization of PSAs of dust throughout the western US not covered in this study.

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## APPENDIX A. SUPPLEMENTARY MATERIAL

Supplementary data associated with this article can be found, in the online version, at http://dx.doi.org/10.1016/j.gca.2017.07.024.

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