Short- and Long-Term Mechanisms for Increasing Inputs of Phosphorus in Mountain Waterbodies of Northeastern Utah, USA

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SHORT- AND LONG-TERM MECHANISMS FOR INCREASING INPUTS OF PHOSPHORUS IN MOUNTAIN WATERBODIES OF NORTHEASTERN UTAH, USA

by

Jessica Scholz

A thesis submitted in partial fulfillment of the requirements for the degree of

MASTER OF SCIENCE in

Watershed Science

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ABSTRACT

Short and Long-term Mechanisms for Increasing Phosphorus Inputs in Mountain Watersheds of Northeastern Utah, USA

by

Jessica Scholz, Master of Science

Utah State University, 2021

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Department: Watershed Sciences

Total phosphorus (TP) concentrations have recently increased in many remote, high-elevation lakes and streams across the western United States and reports of algal blooms in these systems have increased in frequency. In these remote locations, typical anthropogenic TP inputs (e.g., agricultural/urban runoff or wastewater) are implausible explanations for the observed changes. In this study, we investigated alternative mechanisms—atmospheric-P loading and terrestrial-P loading—that may explain increases in P inputs to high-elevation watersheds over the last two decades. Using the Uinta Mountains of northeastern Utah as a case study, we quantified atmospheric P-loading using low-maintenance bulk deposition samplers. Changes in terrestrial P-loading were evaluated through soil leaching and incubation experiments replicating atmospheric acidification and recovery observed in several remote mountain catchments, as well as the changing temperatures attributed to climate change. The proportion of bioavailable-P deposited in dust was determined using sequential chemical extractions. We estimated
bioavailable dust-P loading ranged from 4.9 to 17.7 mg P m$^{-2}$ yr$^{-1}$, and this dust-P flux appears to be one of the most plausible mechanisms for P increases in the Uinta Mountain Range. However, our results revealed that increasing both soil pH and temperatures have the capacity to mobilize significant amounts of P. The mechanisms evaluated in this study can explain the widespread observed increases in P across the Western US, though the mechanisms that dominate in any given region will vary based on local to regional factors.
PUBLIC ABSTRACT

Short and Long-term Mechanisms for Increasing Phosphorus Inputs in Mountain Watersheds of Northeastern Utah, USA

Jessica Scholz

Phosphorus is an essential nutrient that allows life in water to flourish, but changes in phosphorus supplies are not well understood in remote watersheds. In mountain environments, remote watersheds contain critical high-quality water supplies and unique ecosystems, but excess phosphorus can diminish water quality by producing unfavorable aquatic species. Therefore, observed trends of increasing phosphorus concentrations in remote lakes and rivers in the US over the last two decades are a significant concern. Using the Uinta Mountains as a case study, we evaluated three different direct and indirect pathways for phosphorus increases in remote mountain watersheds that may explain observed increases over the last two decades. These mechanisms include atmospheric deposition of dust enriched in phosphorus and changing soil conditions (i.e., increasing soil temperatures and increasing soil pH), leading to greater land exports of phosphorus from runoff or erosion. In the Uinta Mountains, dust had concentrations of phosphorus considered biologically available and could be contributing 4.9 to 17.7 mg P m$^{-2}$ yr$^{-1}$. This likely represents an important source of phosphorus to this area. Also, greater release of SRP from soils incubated at the higher temperatures suggests climate change may further increase phosphorus concentrations in the future. Given the lack of historical acidification in the Uinta mountains, changing pH conditions are not likely to be a factor driving phosphorus increases in this system, but could potentially be in other alpine systems. This study highlights the importance of
identifying local variables in discerning significant sources of phosphorus and disparages a single explanation for national increases in phosphorus in remote systems.
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INTRODUCTION

Nationwide increases in total phosphorus (TP) in surface freshwaters over the last two decades has accelerated the disappearance of oligotrophic (i.e. low-nutrient) status lakes and streams (Stoddard et al. 2016). Oligotrophic lakes and streams have low total phosphorus (TP) concentrations (TP < 10 µg L\(^{-1}\)), constituting sources of high-quality freshwater that require little, if any, treatment prior to human use. In mountainous regions, oligotrophic waterbodies are particularly important. The ecosystem services these aquatic systems provide range from regulating the high-quality water supplies for diverse downstream networks, to providing culturally and spiritually significant sites. However, the quality of oligotrophic waters depends on the nutrient status of their relatively sensitive ecosystems. With slow-weathering bedrock and steep slopes, alpine catchments collect precipitation directly from the atmosphere, creating lakes and rivers with relatively dilute and low-nutrient water. It is likely that even small increases in bioavailable phosphorus (P) within high elevation oligotrophic ecosystems can shift the nutrient concentrations, leading to higher rates of primary productivity and biomass production (Moser et al. 2010; Catalan and Camarero 2014). Furthermore, these shifts in productivity may cascade into lower-elevation ecosystems as nutrients continue to accumulate downstream.

Given the isolation of mountain lakes and streams, the mechanisms for increased P-loading to these ecosystems are unclear. Identifying drivers for high-elevation eutrophication is required to understand which high elevation lakes and streams are more susceptible and the type of management needed to mitigate further increases. With only minor anthropogenic land-use or riparian disturbances in high-elevation catchments, it is
difficult to explain increases in P-inputs with the typical point or nonpoint sources of nutrient pollution (e.g. agricultural run-off or nutrient-rich waste) (Carpenter et al. 1998), and the close association between water chemistry and atmospheric inputs suggests mechanisms are related to large-scale shifts in the atmospheric processes or climate. Considering the importance of mountain ecosystems for maintaining high-quality water, we aim to identify these more elusive mechanisms that may be driving increases in freshwater P-concentrations to relatively isolated mountain watersheds. Below, I will provide an overview of both atmospheric and terrestrial P-loading mechanisms that provide the basis for this study.

Fig. 1 Three pathways for phosphorus inputs in a mountain watershed: Atmospheric deposition, leaching, and erosion. Oval indicates the mechanisms examined in this study, related to atmospheric deposition (both wet and dry) and leaching pathways controlled by soil properties related to acid rain and climate change.
Atmospheric P-loading

Increases in atmospheric deposition of P-rich dust is one mechanism proposed for explaining increases in freshwater P-concentrations within mountain watersheds, particularly mountains close to arid regions such as the southwestern US, the Sahara Desert, the Tibetan plateau, and central Australia (Reche et al. 2009; Brahney et al. 2014; Stoddard et al. 2016). Persistent drought conditions and the onset of major changes in land use within arid and semi-arid climate zones within the last two centuries are linked to increased dust emissions (Belnap and Gillette 1997; Neff et al. 2008; Brahney et al. 2015; Katra et al. 2016). In the southwestern US, atmospheric concentrations of Ca$^{2+}$ (a proxy for dust) provides evidence that dust emissions rates in Utah, Nevada, and California are still increasing. However, in Colorado, dust emissions appear to have decreased since 2010 (Brahney et al. 2013, Clow et al. 2016, NADP, 2020). In many mountain environments in the Western US, geochemical and mineral signatures for long-term atmospheric dust inputs are present in soils and lake sediments, suggesting dust represents an important supply of nutrients to mountain ecosystems (Muhs and Benedict 2006; Neff et al. 2008; Ballantyne et al. 2011; Lawrence et al. 2013; Brahney et al. 2015; Munroe et al. 2015). In fact, in the Sierra Nevada mountains of California, P from atmospheric dust may help sustain nutrient concentrations by outpacing the weathering of phosphate minerals from the granitic bedrock (Homyak et al. 2014; Aciego et al. 2017).

Here we evaluate how increases in the flux of atmospheric dust may contribute to increased P-loading in high elevation sites over shorter time scales.

Despite the evidence for increasing dust deposition at a regional scale, the role of dust as a nutrient vector between source and sink ecosystems is poorly understood (Field
et al. 2010). Historical increases in dust fluxes to freshwater systems correlate to increases in biological activity, changes in lake nutrient ratios, and increased bacterial abundance (Reche et al. 2009; Brahney et al. 2015). However, more studies are needed to understand the variability in dust composition and to quantify the flux of biologically available-P from dust to these catchments.

To better understand the reactivity of P in dust, several studies have separated and quantified different forms of dust-P using sequential extraction and XANES spectroscopy (Chen et al. 2006; Gao et al. 2019). However, it is unclear how dust collection methods and chemical separation methods influence the measurements of P in dry and wet deposition. For instance, inorganic and organic P compounds considered “labile” are readily available for organisms, and these compounds may be actively incorporated into terrestrial and aquatic ecosystems over relatively short time periods (Turner et al. 2005). In contrast, forms of P that are immobile and resistant to chemical or biochemical degradation are less likely to move through the catchment over ecological time scales. Thus these recalcitrant forms of P are unlikely to be contributing to shifts in freshwater ecosystems. Given that P is typically the limiting nutrient in remote, high elevation systems, identifying the biologically available portion of P in dust is essential to understanding the ecological significance of dust to these systems.

**Terrestrial P-loading**

*Effects of Soil pH*

Increasing P fluxes in isolated mountain lakes may also be caused by progressively declining NO$_3^-$ or SO$_4^{2-}$ concentrations in precipitation within previously acidified catchments. The reduced deposition of these acid-forming ions allows base
cations in the soil to replenish, thus increasing the pH of soils and surface waters by allowing base cations to displace the accumulated acidity. This recovery from acidification has been recognized to have ecological implications in areas with extensive historical acidification by shifting aquatic community compositions (Driscoll et al. 2001; Baldigo et al. 2016; Burns et al. 2016) and nutrient cycling (Kopáček et al. 2000; Evans et al. 2001; Driscoll et al. 2003; SanClements et al. 2018). For instance, it is widely accepted that increases in terrestrial DOC loading to surface waters are associated with pH recovery (Monteith et al. 2007; Waller et al. 2012). However, less is known about how pH recovery influences terrestrial loading of other nutrients, including P (Corman et al. 2018), which can represent significant P input into high-elevation lakes over several decades of progressive pH recovery. For instance, increases P-loading is associated with DOC-export and acid recovery in previously acidified alpine lakes in Eastern Europe (Kopáček et al. 2015). However, the different responses to acid recovery across European mountain ranges, suggest that the physical, chemical, and biological catchment characteristics determine the catchments response to acidification and nutrient release (Kopáček et al. 2000, 2019; Kaňa and Kopáček 2006). In cases of heavy acidification, initial pH recovery may cause decreases in P loading by precipitating Al-oxide compounds that bind P within the soil (Huser and Rydin 2005; Kopáček et al. 2005). Thus, more research is needed to understand how different catchment characteristics across high-elevation sites influence changing soil acidity and the subsequent release of P to surface waters.

The connection between soil P release and pH recovery is related to the electric charge of soil particles. As a measure of the activity of H⁺ ions in the soil solution, the
soil pH determines the surface charge of soil particles and thus the binding capacity for both cations and anions in the soil. At lower pH values (high acidity), there are more positively charged sites that can bind the negatively charged P ions in the form of phosphates, including $\text{H}_2\text{PO}_4^{-}$, $\text{HPO}_4^{2-}$, $\text{PO}_4^{3-}$ (Hereafter, \text{PO}_4-P). Following increases in soil pH, more \text{PO}_4-P is thought to desorb, i.e., disassociate, from soil particles due to the decrease in the number of binding sites. Given that organic compounds also have a negative charge, DOC and \text{PO}_4-P are predicted to be co-exported by the same pH mechanism. However, the desorption of \text{PO}_4-P depends on the type and quantity of soil SOM as well as the texture and composition of the soil. For instance, soils with higher metal oxide concentrations and higher clay fractions are more likely to bind more \text{PO}_4-P into highly resistant compounds (Walker and Syers 1976).

Effects of Climate Change

Another mechanism for increases in P fluxes to isolated mountain waterbodies may be climate-driven increases in soil temperature. Longer growing seasons associated with anthropogenic climate change and higher temperatures may accelerate the microbial decomposition of organic material, leading to greater pools of water-soluble organic carbon (Trumbore 1997; Conant et al. 2011) and, consequently, bioavailable-P. However, it is unclear how soil decomposition and nutrient cycling processes in high elevation regions may respond to the increases in temperature and extended growing seasons.

In alpine regions, the availability of P in soils is primarily controlled by the decomposition and biogeochemical cycling of soil organic matter (SOM) (Weintraub 2011). With relatively small inputs of P from mineral weathering or atmospheric deposition, organic P compounds are typically conservatively cycled and mineralized into
available forms by soil microbes. Thus, the amount of water-soluble P in the soil that can be leached by precipitation is relatively small. However, increased decomposition of SOM with higher temperatures may be releasing more dissolved organic carbon (DOC) (Christ and David 1996; Evans et al. 2005) and presumably organic-P into the soil solution at a rate higher than biological uptake. However, the sensitivity of different pools of SOM to increases in temperature regions is debated, and relatively few studies have examined the effects of warming on the P fluxes from soils (Conant et al. 2011).

In this study, we used contemporary soil and dust sampling as well as experimental analysis to evaluate the relative significance of these three competing hypotheses for increases in P within isolated, high-elevation lakes and streams.

**Hypothesis 1**: Increased fluxes of atmospherically deposited “dust” contains bioavailable-P contributing to P concentrations and exports in alpine and subalpine waterbodies.

**Hypothesis 2**: Rising soil-pH from the reduced acidity of precipitation is increasing the amount of P leached from catchment soils.

**Hypothesis 3**: Progressive warming of soils is increasing the amount of P leached from catchment soils.

Our study examines P-inputs in four remote, high-elevation catchments in northeastern Utah, USA, with a focus on high elevation sites in the Uinta Mountains (hereafter, Uintas). The Uintas were the primary focus of this study for several reasons: 1) the Uintas contain high-elevation streams and lakes included in the EPA sample set of minimally disturbed sites that show rising TP concentrations between surveys conducted between 2000 and 2014 (Stoddard et al. 2016); 2) The Uintas have relatively uniform
quartzite bedrock composition, allowing us to minimize differences in soil parent material and compare differences in the dust fluxes and soil PO₄-P leaching responses between E and W catchments; 3) Previous characterizations of soil pedons from high-elevation sites in the Uinta Mountains allowed us to predict whether changes in soil pH have occurred by comparing historical and current data (Bockheim 1997, Bockheim et al. 2000, Munroe 2007). 4) Data for long-term soil and air temperature, as well as dust phosphorus fluxes in high elevation sites, are available for the Uinta Mountains (Reynolds et al. 2010). Additionally, it is important to note the Uintas are bordered to the south by the Uintah Basin, a relatively arid area with extensive oil and gas extraction operations, and bordered to the east by a large-scale phosphate mine, making it likely that the Uinta Mountains have received fluctuating levels of acid deposition, as well as dust deposition, correlated to the production and expansion of these industries over the last few decades.

The purpose of this study is to provide a first attempt at quantifying the relative importance of three different mechanisms for increased P-fluxes within this geographic area. Studies with multiple hypotheses are increasingly valuable as a means of eliminating cognitive biases towards a single explanation (Betini et al. 2017). To test Hypothesis 1, we collected and analyzed P fractions in dust; to test Hypothesis 2, we measured the SRP leached from soils along an imposed pH gradient; to test Hypothesis 3, we measured SRP leached from soils along a temperature gradient. We anticipate the results from this study will help direct future research on P cycles and land-water interactions in high-elevation systems.
Fig. 2  Northeastern Utah study region showing (a) Uinta sites and other high-elevation sites used for comparisons: Bear River Mountains and Wasatch Mountains. TG- Tony Grove site, WP- Willard Peak Site, West-U – West Uinta Site (Provo River Drainage basin), East-U – East Uinta site (Chepeta Basin) (b) drainage basins in the Uinta Mountains investigated in this study. Points mark the West and East drainage basins selected in the Uinta mountains. (c) West-U - Provo drainage basin (d) East-U Chepeta basin.
METHODS

Site Description

The northeastern region of Utah (Fig. 2) contains mountain ranges of varying geologic age and parent material. The Uinta mountains, located between 109° and 111° W and 40° to 41° N, are some of the oldest mountains in the Western US, composed of Precambrian (greater than ~ 500 million years old) quartzite and arkose sandstones that were originally deposited during the Neoproterozoic (1000-700 Ma) (Condie et al. 2001). Unlike most mountain ranges in the Western US, the Uintas formed much earlier and are oriented E-W instead of N-S. This orientation creates a relatively unique system with a precipitation gradient decreasing from W to E.

In the Uintas, dust and soil samples for P-analysis were collected from both a west and east-central drainage basin (Fig. 2b). Both sites were selected because they were accessible by foot and within 5 miles of roads. Both sites also have SNOTEL stations (Trial Lake and Chepeta), allowing us to evaluate potential differences in precipitation, air temperature, and soil temperature between the sites.

In the western Uintas, samples were collected in the Provo River drainage basin within the Wall Lake (WL) catchment (Fig. 2c), one of the headwaters for the Provo River. This basin within the Uinta-Wasatch-Cache National Forest contains several campgrounds along the shores of south-neighboring Trial Lake. Hiking trails from these campgrounds lead to Wall Lake and higher elevation lakes within the drainage catchment. In the eastern Uintas, samples were collected in the Chepeta basin (Fig. 2d). The lakes within the Chepeta basin drain into the Whiterocks River. This basin is in the Ashley National Forest, and it is more isolated than the Provo River basin.
In addition to the Uinta Mountains, soil samples were collected from the Wasatch Range and the Bear River Range, located west and northwest of the Uinta Mountains (Fig. 2a). These sites were chosen because of their accessibility and proximity to headwaters that drain to springs accessed by nearby cities.

The Wasatch Mountains are located west of the Uinta Mountains and span the eastern side of Utah Valley in Northern Utah, one of the most densely populated regions of Utah. The geology of the Wasatch Range consists of limestone, dolostone and quartzite deposits, with metamorphosed shales and felsic igneous deposits. The site selected in the Wasatch Range in the Willard Peak (WP) basin contained primarily of weakly metamorphosed shale. At Willard Peak, there is a single lake draining into Willard creek and the Willard City Spring.

The Bear River Range is often considered part of the Wasatch Range (Dastrup et al. 2018), but here it will be referred to separately as the Bear River Range due to its relatively distinct geologic composition and geographic separation. The bedrock of the Bear River range is composed primarily of limestone and dolostone interbedded with quartzite. The site selected for soil sampling is within the Tony Grove (TG) catchment, which drains into the Logan River in northern Utah.

**Dust Sampling and Analysis**

*Dust Sampler Design*

Dust samplers for this experiment were built and tested as low-maintenance samplers to collect both dry and dissolved deposition from wilderness areas in the Uinta Mountains (Fig. 3). Previous dust studies in the Uintas have used passive marble dust traps placed on the ground (Munroe 2014a), and similar type marble traps. We were
interested in analyzing P in both dry and wet deposition; therefore, the samplers were modeled after bulk deposition samplers and resin ion exchange columns used to measure ion deposition in throughfall and precipitation (Fenn et al. 2003).

Precipitation rates average 770 mm/yr (Chepeta) and 1000 mm/year (Trial Lake, SNOTEL), therefore dust samplers in this study needed to be able to collect dust and ions without overflowing. We addressed this problem by creating a sampler that traps dust and dissolved ions while allowing water to drain out. Each sampler consisted of a large funnel attached to a column filled with ion-exchange resin. A filter was placed at the base of the funnel to trap the dust, while small holes at the base of the resin-filled column allowed precipitation to drain. The resin was placed in the column to bind the dissolved ions in the precipitation. To avoid contamination from wildlife, triplicate sets of samplers were mounted 1.5 meters off the ground onto a pole set up in open meadows. Dust samplers

![Fig. 3 Resin column dust samplers used in this study. Image of samplers at West site, samplers were placed about 10 meters away from standing trees. Given the hiking traffic through this area, we avoided open areas that would be too conspicuous.](image-url)
were retrieved after three months at the end of summer to prevent possible disturbances to the samplers from snowfall. To capture both negative and positively charged ions, two sets of triplicated samplers, one filled with Na\(^+\) resin and the other filled with Cl\(-\) resin were placed at each site. Prior to installation, all materials were acid washed with 0.1M HCL and triple rinsed with milli-Q water.

Dust Sampling and Processing

In the Provo River drainage Basin (West site) and the Chepeta Basin (East site), six dust samplers were deployed at each site, three containing anion resin, and three containing cation resin (Fig. 3). In the West site, dust samplers were left out July 2- Oct. 19 (109 days) and in the East site, samplers were left out June 6- Oct. 19 (138 days).

We collected and analyzed a total of 6 replicate samples of dust from the East Uinta Site, and 8 replicate samples of dust from the West Uinta site, deposited during the Summer of 2018. Upon collection, three of the six funnel samplers deployed in the East Uinta site were found upright. To minimize error from local soil contamination, we only analyzed samples collected on those 3 upright samplers. Similarly, at the West Uinta site, four of the six samplers were found upright, and thus, we only analyzed samples from those four upright samplers. Filters collecting dust were removed from base of the funnel and placed in sterile polyethylene bags. Funnels and marbles were also removed and stored in separate labeled sterile polyethylene bags. The resin column below the funnel were capped with a sterile lid. The samplers, along with all filter and resin samples, were taken back to Utah State University for processing.

To ensure enough replicates for the sequential extraction, we collected additional dust by rinsing the funnel and marble with Milli-Q water, and then filtering the solution
through a 0.45µm poly filter to isolate the solid dust particles. These additional dust samples were collected from all 3 upright samplers from the East Uinta site, and all 4 upright samplers from the West Uinta site.

To supplement the summer dust samples collected from the Uintas sites and to compare seasonal differences in P concentrations, we analyzed dust-on-snow samples collected in two sites located on the Central and East side of the Uintas (Lakefork and Grizzly Ridge). To collect both wet and dry deposition from the snowpack, depth-integrated snow samples were collected into clean Teflon bags at 6 locations using USGS protocols (Ingersoll et al. 2002) and then sent to Utah State University for processing. Snow samples were allowed to melt in the Teflon bags at room temperature prior to filtration through pre-weighted 0.45 µm PES filters. Each bag was rinsed three times with DI water and filtered to maximize the recovery of materials from the Teflon bag. After filtration, the PES filters with the dust were dried in a desiccator for 24 hours and weighted on Metler-Toledo Excellence XS105DU Analytical Balance and Anti-Static Kit.

*Phosphorus Sequential Extraction*

To identify the amount of bioavailable P in dust deposited in the Uintas, we used a sequential extraction procedure modeled after the SEDEX method for P speciation in sediments (Ruttenberg 1992), and the “Hedley method” commonly used for P speciation in soils (Hedley 1982), to quantify five increasingly resistant forms of P (Fig. 4).

Sequential extraction methods are often used on soil and sediments samples to separate P forms based on their reactivity. Previous studies analyzing P speciation in dust have used modifications of the SEDEX method (Chen et al. 2006; Zhang et al. 2018),
Fig. 4 Sequential phosphorus extraction method used for separating and quantifying the different forms of phosphorus in dust. The forms of P extracted, from least to most reactive include: (i) water-soluble P (DI-P) (ii) exchangeable P (Bicarb-P, extracted with 0.5 M sodium bicarbonate solution at pH 8) (iii) Fe-Al associated P and organic-P (NaOH-P, extracted with 0.1 M sodium hydroxide solution), (iv) authigenic apatite P (Acet-P, extracted with an acetic acid-sodium...
developed originally for marine sediments, using the rationale that dust will eventually end up as components of lake or ocean sediments. However, on shorter timescales within catchments, dust is deposited directly onto the soil and water surfaces and incorporated into biologically active systems. For these reasons, we combined SEDEX with Hedley’s soil-P fractionation, a common fractionation method used to quantify the bioavailable forms of phosphorus in soils (Hedley 1982).

The following operationally defined phosphorus fractions were quantified: (i) water-soluble P (DI-P) (ii) exchangeable P (Bicarb-P, extracted with 0.5 M sodium bicarbonate solution at pH 8) (iii) Fe-Al associated P and organic-P (NaOH-P, extracted with 0.1 M sodium hydroxide solution), (iv) authigenic apatite P (Acet-P, extracted with an acetic acid-sodium acetate solution pH 4), and (v) detrital P (total digestion with nitric acid HNO₃). For this study, DI-P, Bicarb-P and NaOH-P were considered bioavailable. DI-P and Bicarb-P are widely accepted as readily available forms of P for microorganisms in soils and waters (Hedley 1982; Levy and Schlesinger 1999). We also included NaOH-P because this fraction has correlates well with algal-available P forms in bioassay studies (Sonszogni et al. 1982; Dorich et al. 1985).

Prior to the sequential extraction, filters with dust from the bulk were weighed pre-and post-filtration to determine dust mass. For each extraction step, 10 mL of the extractant solution was added to all 16 dust-on-filter samples, which were cut into pieces to fit in 50 mL centrifuge tubes. One blank was also carried through all the extraction steps. Samples were shaken for 16 hours prior to centrifuging at 3500 RMP for 10 minutes. The supernatant solution for each sample was decanted and placed into separate acid-washed containers for P analysis. Between extraction steps, 10 mL milli-Q water
was added to each sample and shaken for 4 hours to help rinse the previous extraction solution and residual P from the dust samples. This supernatant solution from the milli-Q water rinse was added to the previous extractant solution. Samples were dried overnight before adding the next extractant in the sequence. After all extraction steps were completed, the remaining dust residue was analyzed for elemental P concentrations using an inductively coupled plasma-mass spectrometer (Agilent 7700x ICP-MS) at the Utah State University Water Lab. Prior to ICP-MS analysis, the remaining dust was digested using nitric acid to measure TP. Subsamples of the digestion were filtered through 0.2 µm filters and acidified with nitric acid by 1% of the sample volume to preserve the elements in solution.

The resin within the Uintas dust samplers’ columns was analyzed for PO₄³⁻ ions that we report separately from the dust result as “dissolved-P”. Dissolved-P represents P from precipitation and leached from of dust prior to collection. Of the three samplers containing positively charged resin deployed at each site, one from the East and three from the West, were successfully retrieved for analysis. To extract the for PO₄³⁻ ions, 200 mL of 1M HCl was added to each of the 4 resin samples. The resin was rinsed twice with 100 mL of DI water. The HCl and DI water was combined and analyzed for PO₄³⁻ using a Lachat 8500 Flow Injection Analyzer.

Differences in bioavailable-P, recalcitrant-P, and TP concentrations in dust were evaluated between the East and West sites using t-test. However, with only 1 resin sample from the East Site, differences in dissolved-P between the East and West sites could not be statistically evaluated.
**Table 1** Soil Sampling Summary for intact soil core samples used to test the temperature hypothesis (top) and bulk soil samples used to test the pH hypothesis (bottom)

<table>
<thead>
<tr>
<th>Vegetation</th>
<th>Core soil samples</th>
<th>Bulk soil samples</th>
</tr>
</thead>
<tbody>
<tr>
<td>Pine</td>
<td>16</td>
<td>10</td>
</tr>
<tr>
<td>Shrub/grass</td>
<td>12</td>
<td>14</td>
</tr>
<tr>
<td>Total</td>
<td>28</td>
<td>24</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Vegetation</th>
<th>Wall Lake (WL)</th>
<th>Taylor Lake (TL)</th>
<th>Hiddlen Lake (HL)</th>
<th>Willard Peak (WP)</th>
<th>Tony Grove (TG)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Pine</td>
<td>9</td>
<td>9</td>
<td>9</td>
<td>--</td>
<td>--</td>
</tr>
<tr>
<td>Shrub/grass</td>
<td>9</td>
<td>9</td>
<td>9</td>
<td>--</td>
<td>--</td>
</tr>
<tr>
<td>Total</td>
<td>18</td>
<td>18</td>
<td>18</td>
<td>--</td>
<td>--</td>
</tr>
</tbody>
</table>

**Soil Sampling and Processing**

Bulk soil samples were collected in the Provo River Drainage Basin and the Chepeta Basin in the Spring/Summer of 2018 for the pH experiment. One catchment was selected in the Provo River drainage basin, and two catchments in the Chepeta Basin. We intended to collect samples from both an alpine and subalpine lake catchment in each basin, but in the Provo River drainage site we only sampled a subalpine catchment due to inaccessible terrain. Soils types were grouped by the general vegetation: pine and grass/shrubs. From the two vegetation groups, we randomly selected three sites for sampling with three replicate samples composited from each plot, creating a total of 18 soil samples (Table 1).

The bulk soil samples were collected by first removing the top layer of vegetation and then extracting the O and A layers (2-10cm) of soil using a soil probe. Soils were stored in polyethylene bags and kept on ice until returning to the Utah State University
Environmental Biogeochemistry Lab, where they were refrigerated at 4 °C prior to analysis. Soils were dried at 60 °C for 24 hours and then passed through a 2 mm sieve to remove rocks and vegetation. For each catchment site, equal weights of the three samples from each vegetation type were combined, creating one bulk pine and one bulk grass/shrub sample from each site, producing a total of six bulk soil samples. Bulking soil samples was more feasible for the pH experiment because it reduced the number of replicates needed for the seven pH treatments for each soil type.

Intact soil samples for the temperature experiments were collected in September 2018 from three high elevation catchments in Northeastern Utah: Uinta Mountains, Bear River Range, and Wasatch Range. From each catchment selected, 9-15 soils (depending on the size of the catchment) from Pine and Shrub/Grass samples were randomly sampled. Intact soils cores were collected in PVC pipes 2.5 cm diameter and 10 cm length. Pipes were hammered into the ground until at least 2/3 full of soil (average soil depth in pipe: 6.3 ± 1.7 cm). The pipe was withdrawn carefully and then wrapped in plastic film to preserve the soil structure. Samples were kept on ice until returning samples to the lab where they were stored at 4°C until analysis. This sampling method was chosen to preserve the natural state and structure of the soil. Previous studies have shown that disturbance and drying the soil can alter microbial activity and rates of nutrient mineralization (Condron and Newman 2011).

**Soil pH Experiment**

Changes in soil pH within the Uintas were evaluated by comparing past soil pH data with the soil pH data collected in this study. Similar soil types were selected from
Bockheim (1997) and Bockheim and Munroe (2000), and Munroe (2007) to represent historical pH values.

The potential for increases in soil pH to increase P concentrations to high-elevation lakes was evaluated experimentally by subsequently leaching soils after altering the pH. To determine the effects of pH on the amount of soluble reactive P (SRP) released from soils, experiments were conducted for each of the six different bulk soil samples. Methods for the pH experiment are modeled after Kopáček et al. (2015). In these experiments, soil samples were adjusted to seven different pH levels (4.0, 4.5, 5.0, 5.5, 6.0, 6.5 and 7) in triplicate, and the soil leachate was measured for SRP. Experiments were carried out separately for each soil type to avoid contamination from the pH probe. Three one-gram subsamples were used for each different pH treatment, creating a total of 21 samples per soil type. After adding 30 mL of DI water to each sample, the soil-water solutions were adjusted to the desired pH level using drops of 0.1 M HCl and 0.1 M NaOH. Samples were then placed in a Thermo Scientific MaxQ 4000 Orbital Shaker adjusted at 300 RPM for 24 hours. The pH of the samples was adjusted every two hours for the first 12 hours, and then every six hours for the remaining 12 hours. After the samples had been shaken for 24 hours and were within 0.05 of the desired pH, they were placed in a centrifuge for 10 minutes. The supernatant solution was then filtered using 0.45 um fiberglass filters and placed into a separate container for SRP analysis using the molybdate method (Murphy and Riley 1962) on a SpectroMax M2E microplate and cuvette reader.

The effect of soil pH and soil type on the amount of SRP released from the soil was evaluated using a standard two-way ANOVA. To help represent the experimental
results for each soil type, 95% confidence intervals were created using a Monte Carlo simulation. We simulated a dataset for each soil type using the mean and standard deviation of the replicate leachate samples carried through the experiment. A normal distribution (n = 100) was generated using the sample mean and standard deviation. Using the simulated data, 95% confidence intervals were created for each plot. The amount of P released for a m² plot of soil was estimated by assuming a soil A horizon depth of 5 cm and a soil density of 1 g cm⁻³.

**Soil Temperature Experiment**

To determine how changes in soil temperature within high elevation catchments influence SRP release from soils, we conducted temperature experiments in growth chambers (Conviron Model A1000) with three different incubation treatments representing historical (C), current (T₁), and our future-projected increases in soil temperatures (T₂) over a simulated growing season. The simulated growth seasons for each treatment were produced from average historical soil temperature data from the Snow Telemetry (SNOTEL) network of high elevation sites in the Uinta Mountains. Each day in the simulated growing season represented the average temperatures over a full week during the growing season. The rationale for simulating growing season temperatures instead of incubating at constant temperatures is based on the recognition that certain microbes are active only during particular soil temperatures or times of the day (Koch et al. 2007). Selecting one average temperature would not reflect the actual increase and decrease of temperatures and hours of sunlight that occur in the natural environment.
Table 2 Summary of Incubation Treatments

<table>
<thead>
<tr>
<th>Treatment</th>
<th>Average Temperature (°C)</th>
<th>Duration of Treatment (days)</th>
</tr>
</thead>
<tbody>
<tr>
<td>C</td>
<td>8.1</td>
<td>12</td>
</tr>
<tr>
<td>$T_1$</td>
<td>10.6</td>
<td>13</td>
</tr>
<tr>
<td>$T_2$</td>
<td>12.4</td>
<td>14</td>
</tr>
</tbody>
</table>

The average temperatures and duration of the three incubation treatments are summarized in Table 2. For temperatures in the C treatment, we averaged weekly temperatures during the weeks in the year when temperatures exceeded 5°C from 2001-2005 for all high elevation sites in the Uinta Mountains. A minimum of 5 °C was selected because it was the minimum temperature for the incubators used in our study. Similarly, for the $T_1$ treatment, we averaged temperatures during the weeks when temperatures exceeded 5 °C between 2013 and 2017. For $T_2$ treatment, we added 2 °C to the $T_1$ treatment to represent increases in temperature that are predicted to occur with climate change. The decision to simulate a condensed growing season, where one day represented the average temperatures over a week (instead of using the length of the growing season) was made to make the incubation length more feasible. Furthermore, we did not believe a longer incubation period would more closely replicate natural conditions, given that new organic matter would be continuously added to the soils in the natural environment.

Using the temperature data for the Uinta sites, we were able to determine which sites have experienced significant increases in the number of days where temperature exceeds 5°C. (see Appendix B for full range of temperatures used over the incubation period for each treatment). The water content of the soils was adjusted to approximately 50% prior to the incubation. The density of the soils was determined using soil collected
in the sediment cores. The cores were covered in plastic with small holes to allow gas
eye exchange with minimal water loss. However, some water loss did occur, but due to the
short duration of the experiment, the water loss was presumed to have little impact on the
results. After the incubation period, a 1-g subsample from each column was leached with
30 mL of water. The amount of P released for a square plot of soil was estimated by
assuming a soil A horizon depth of 5 cm and a soil density of 1 g cm$^{-3}$.

Differences in SRP and TP released from soils between treatments (C, T$_1$, T$_2$)
were analyzed using the Kruskal-Wallis test as a nonparametric alternative for ANOVA.
Because of unequal variances and sample sizes, the Games-Howell test was used to
determine the pairwise differences in SRP and TP released between treatment.
RESULTS

Uinta Dust

Sequential Extraction of Phosphorus from Dust

Dry deposition TP concentrations (as the sum of DI-P, Bicarb-P, NaOH-P, Acet-P, and Residual-P fractions) were 2.1 mg P g⁻¹ dust, with similar concentrations between the East and West samples at 1.96 and 2.20 mg P g⁻¹ dust, respectively (t-test, p = 0.30) (Fig. 5a).

For the winter deposition, dust-on-snow samples from Lake Fork and Grizzly Ridge sites were combined for P analysis. The TP concentration of these samples is 1.42 mg P g⁻¹ dust, which is less than TP concentrations in the summer deposition (Fig. 6). The average TP concentration between summer and winter deposition is 1.73 mg P g⁻¹, which falls between previous Uinta dust measurements of P in coarse and fine dust fractions, at 0.24 and 4.0 mg P g⁻¹ dust, respectively (Munroe 2015).

**Fig. 5** Distribution of P concentrations in East and West Uinta summer dust samples, showing (a) total phosphorus (TP), (b) bioavailable phosphorus, and (c) recalcitrant phosphorus concentrations (mg P g⁻¹)
Uinta dust TP concentrations are greater than the global average of 1.09 ±0.14 mg P g\(^{-1}\) dust, reported by Lawrence and Neff (2008). Compared to other alpine sites, concentrations of TP measured in the winter Uinta dust (dust-on-snow) are higher than dust-on-snow TP measurements spanning the Colorado Rockies, at 0.56–0.96 mg P g\(^{-1}\) dust (Lawrence et al. 2010; Zhang et al. 2018). TP concentrations in summer Uinta dust are greater than summer dust measurements from the Sierra Nevada range in California, 1.5 ± 0.2 mg P g\(^{-1}\) dust (Aciego et al. 2017), but were less than those reported in the Wind River Mountains, WY, 3.0 ± 0.15 mg g\(^{-1}\) dust (Brahney et al. 2014) and southeastern Australia at 4.3 ± 1.81 mg g\(^{-1}\) (Leys and McTainsh 1999).

**Fig. 6** Comparison of phosphorus fractions between Summer and Winter dust from the Uinta Mountains. No error bars are shown for the winter deposition because there were no replicates.
For summer deposition, the average P concentrations considered bioavailable (DI-P, Bicarb-P, and NaOH-P) is 0.94 mg P g\(^{-1}\), also with similar concentrations between the West and East, at 0.96 and 0.92 mg P g\(^{-1}\) dust, respectively (t-test, \(p = 0.88\)), representing 42-50% of the TP in dust. The portion of bioavailable-P for the winter dust is smaller than summer measurements, at 0.46 mg P g\(^{-1}\) and represents a smaller portion of TP, at 32%. In summer dust, the recalcitrant (Acet-P, Resid-P) fraction appears to be slightly greater in the West Uinta dust at 1.28 mg P g\(^{-1}\) compared to East Uinta dust at 1.00 mg P g\(^{-1}\), but differences were not statistically significant (t-test, \(p = 0.15\)) (Fig. 5b, 5c, 6). The recalcitrant-P concentration for the winter deposition is 0.96 µg P g\(^{-1}\) dust, which is similar to the summer dust measurements (Fig. 6).

**Dust-Phosphorus Flux**

P flux values were calculated using dry deposition rates in the Uinta mountains reported by Munroe (2014b) and Brahney (2019, 2020), which are 3.6, 6.6, and 5.9 g dust m\(^{-2}\) yr\(^{-1}\), respectively (Table 3). The dust deposition rate reported by Munroe (2014) is the average annual deposition over 2 years (2012-2013) of four sites representing the North, East, West, and South regions, which ranged from 2.7-5.5 g dust m\(^{-2}\) yr\(^{-1}\). The dust deposition rates reported by Brahney (2019, 2020) were determined from dust on-snow collected in the East Uinta region and wet and dry particulate deposition collected through the NADP network. These dust deposition rates are similar to other mountain sites in the Western US, including the Wind River mountains, at \(~1-4\) g m\(^{-2}\) yr\(^{-1}\) (Brahney et al. 2014), the Colorado front range at 5.9 ±0.2 g m\(^{-2}\) yr\(^{-1}\) (Heindel et al. 2020), the San Juan Mountains at \(~5-10\) g m\(^{-2}\) yr\(^{-1}\) (Lawrence et al. 2010), and the Sierra Nevada mountains, CA 3-36, average \(~10.6\) g m\(^{-2}\) yr\(^{-1}\) (Aciego et al. 2017).
Using these dry deposition rates, we estimated the flux of bioavailable-P from dry deposition to be 8.8 to 17, with an average of 14 µg P m\(^{-2}\) day\(^{-1}\) in the summer and 7.9 µg P m\(^{-2}\) day\(^{-1}\) in the winter. The TP flux is approximately 29.0 µg P m\(^{-2}\) day\(^{-1}\) in the summer and 24.3 µg P m\(^{-2}\) day\(^{-1}\) in the winter. Scaled to an annual rate, assuming a winter season lasting 8 months, and summer lasting 4 months, we predict an annual flux of bioavailable P to be 4.6 mg P dust m\(^{-2}\) yr\(^{-1}\) and 9.4 mg P dust m\(^{-2}\) yr\(^{-1}\) for TP.

Table 3  Uinta dust fluxes and estimated P fluxes

<table>
<thead>
<tr>
<th>Source</th>
<th>Annual dust deposition (g dust yr(^{-1}))</th>
<th>Bioavailable-P (mg P m(^{-2}) yr(^{-1}))</th>
<th>Recalcitrant-P (mg P m(^{-2}) yr(^{-1}))</th>
<th>Total P (mg P m(^{-2}) yr(^{-1}))</th>
</tr>
</thead>
<tbody>
<tr>
<td>Munroe 2014</td>
<td>3.6</td>
<td>3.5</td>
<td>4.2</td>
<td>7.7</td>
</tr>
<tr>
<td>Brahney 2019</td>
<td>6.6</td>
<td>6.4</td>
<td>7.9</td>
<td>14.3</td>
</tr>
<tr>
<td>Brahney 2020</td>
<td>5.9</td>
<td>5.8</td>
<td>7.0</td>
<td>12.8</td>
</tr>
<tr>
<td>Average</td>
<td>5.4</td>
<td>5.2</td>
<td>6.4</td>
<td>11.6</td>
</tr>
</tbody>
</table>

For the summer deposition, we were also able to retrieve measurements of P as PO\(_4^{3-}\) from one resin column from the West and three from the East Uinta site (measured on the Lachat 8500 Flow Injection Analyzer). This dissolved-P, representing PO\(_4^{3-}\) dissolved in (or leached by) precipitation over the sampling period, here is reported separately from bioavailable-P because it may overestimate P from external dust sources. Over the sampling period, the deposition rate was 92.6 µg P day\(^{-1}\) m\(^{-2}\) in the West and 125.9 ± 87.0 µg P m\(^{-2}\) day\(^{-1}\) in the East, which are an order of magnitude greater than our predicted fluxes of bioavailable-P from dust. We presume some of this P is from local contamination of organic matter, which would be consistent with the large disparity between the three resin sample measurements (Table 4). Samplers were placed in subalpine catchments with stands of coniferous trees nearby, and several pieces of pine and bark were found inside the samplers. Though these local organic contaminants were
removed before dust analysis, this organic matter likely leached nutrients that bound to the resin.

**Table 4**  P as orthophosphate (PO$_4$\textsuperscript{3-}) extracted from resin columns in Uinta dust samplers. Funnel area used to calculate daily (µg P m$^{-2}$ day$^{-1}$) and annual fluxes (mg P yr$^{-1}$ m$^{-2}$)

<table>
<thead>
<tr>
<th>Site</th>
<th>Total P-PO$_4$\textsuperscript{3-} (µg)</th>
<th>Sampling Period</th>
<th>Sampling Duration (days)</th>
<th>P-PO$_4$\textsuperscript{3-} deposition (µg P m$^{-2}$ day$^{-1}$)</th>
<th>Annual P-PO$_4$\textsuperscript{3-} deposition (mg P yr$^{-1}$ m$^{-2}$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>West (n=1)</td>
<td>544$^*$</td>
<td>July 2 - Oct 19</td>
<td>109</td>
<td>92.6$^*$</td>
<td>33.7$^*$</td>
</tr>
<tr>
<td>East (n=3)</td>
<td>944 ± 644</td>
<td>June 6- Oct 19</td>
<td>138</td>
<td>125.9 ± 87.0</td>
<td>46.2 ± 31.5</td>
</tr>
</tbody>
</table>

**Influence of pH on Phosphate Desorption**

**Soil pH Measurements**

Uinta soils are relatively acidic, with pH values ranging from 4.6 to 5.5. The soils from the two vegetation types (Pine and Grass/Shrub) showed no significant differences in soil pH (t-test, p = 0.11) across all sites (see Appendix A for comparisons of soil pH), but the individual sites in the East and West Uintas had unique pH values (ANOVA, p = 0.003) with the subalpine WL catchment in the East Uintas having the highest pH (5.3 ± 0.1), followed by the alpine TL catchment in the West Uintas (5.2 ± 0.2), and the subalpine HL in the West Uintas (4.8 ± 0.1).

The average “historic” soil pH for Uinta soil, between 1997 and 2007, was 5.1 ± 0.1 for the grass/shrub soils (n = 24), and 5.1 ± 0.2 for pine soils (n = 8). All “historic” soils were sampled within the Chepeta Basin, the same drainage basin our East Uinta sites were located, therefore we only compared these values to pH measurements collected from East Uinta soils (TL and HL). The average “current” soil pH from this study was 5.0 ± 0.1 for the grass/shrub soils, and 5.1 ± 0.3 for the pine soils. We did not
detect significant differences in soil pH between historical and current soil pH data in the East Uintas for either shrub or pine soils (t-test, p =0.65) (Fig. 7). No historical data was available for the catchments soil in the Western Uintas.

**Fig. 7** Comparison of historical soil pH data from Bockheim (1997) and Bockheim and Munroe (2000), and Munroe (2007) to soil pH measured in this study.

**Manipulated Soil pH Experiment**

For each site (HL, WL, and TL) results for the average SRP released at each adjusted pH (4, 4.5, 5, 5.5, 6, 6.5, 7) values are listed in Appendix B. SRP values in leachate were normalized by the mass of soil used, and the units represent the amount of SRP released per gram of soil. SRP measurements of the leachate solutions that were below the detection limit of the colorimetric analysis (DL =10 µg P/L) are written as <0.3 µg P g⁻¹ soil and were omitted from the model. Across all soil sites and vegetation types, we found that soils adjusted to the to higher pH values (5.5, 6, 6.5, 7) released the same
or more SRP than soils adjusted to lower pH values (4, 4.5, 5), apart from grass/shrub soils from HL in the East Uinta catchment (Fig. 9b). For the HL grass/shrub soil type, the average amount of SRP appears higher at pH of 5 than soils adjusted to pH of 5.5. It is unclear why this occurred, but we note that it was difficult to stabilize the pH at higher values over the 24-hour period for this soil type, and these soils have high organic matter content (appendix A). Nevertheless, for the remaining soils, we observed a trend of increasing SRP with increasing soil pH observed in soils from the Tatra mountains similarly acidified and alkalinized. (Kopáček et al. 2015) (see supplementary Fig. S2).

Using soil pH and site as explanatory variables, leached SRP concentrations from all data were put into an exponential model (Fig. 8, see Appendix A for model diagnostic plots and model information).

\[
SRP \text{ ug P g}^{-1} = 0.0023 \times 10^{1.1 \times \text{pH}}
\]

The responses of the individual soils can be seen in Fig. 9, with 95% confidence intervals created using Monte Carlo simulations. TL soils, from the highest elevation catchment released less SRP at high pH values when compared to the lower elevation catchments – WL and HL. The ambiguous effects of vegetation type are apparent in these plots; for both WL and TL catchments, pine soils appear to release more SRP than grass/shrub soils, but in the HL catchment Grass/shrubs soils released more SRP compared to Pine soils.

Extrapolated relationships between changes in pH and the amount of SRP released from soils are shown in Fig. 10. This model was generated to compare the effects of pH to the other two mechanisms in this study.
Fig. 8 Relationship between adjusted soil pH and amount of soluble reactive phosphorus (SRP) released in units of µg P g⁻¹ soil for (a) Taylor Lake soils (TL) (b) Wall Lake (WL) and (c) Hidden Lake Soils (HL) soils for different vegetation types. Both (a) and (c) are sites on the Eastern region of the Uintas (b) is from the Western region. Results are displayed with 95% confidence interval generated from Monte Carlo simulated data. (d) Image of test tubes containing leachate from HL soils showing the change in color with increasing pH. (e) SRP plotted for pH, all 6 soil types are included Model for SRP as a function of pH. Note the log-transformed y-axis.
Soil Temperature Experiment

Across all sites, average concentrations of SRP leached were greater from soils incubated at conditions defined in the “current” temperature treatment (T1) compared “historical” (C) treatments by 2.6 ± 1.9 µg P g soil⁻¹ (p = 0.006, Games-Howell test) and greater from soils incubated at the “future” temperature (T2) treatments compared the C treatment by 4.3 ± 2.9 µg P g soil⁻¹ (p = 0.002) (Fig. 11).

To conceptualize this value, we converted the amount of SRP leached per square meter area of a catchment, assuming a soil density of 1.0 g cm⁻³ and organic soil horizon depth of 5 cm. Between C and T1, the difference in SRP release is 130 ± 95 mg SRP m⁻², and between C and T2, it is 215 ± 145 mg SRP m⁻². Because pine soils were shorter on average compared to grass soils, we assumed an average depth of 5 cm, representing the average A horizon depths for the two vegetation types in this study. However, the rates and depths of precipitation infiltration will depend on the intensity and duration of precipitation, slope of land, and soil characteristics. Thus, these estimates represent potential increases in SRP that may occur with elevated temperatures.

At individual sites, significant differences in SRP released were detected only in the Wasatch and Bear River soils, between the C and T2 treatments. In the Wasatch soils, the SRP increased by 7.1 ± 5.0 (p = 0.01), and by 10.0 ± 9.1 µg P g⁻¹ soil in the Bear River site (p = 0.04). These increases translate to 355 ± 250 and 500 ± 455 mg SRP released per square meter, respectively. The magnitude of P released from East Uinta soil was considerably less than the three other sites. The SRP released from the different soil sites and treatments are shown in Table 5.
Fig. 9 Phosphorus leached following the three temperature treatments (C, T₁, T₂) (a) SRP (µg P g-soil⁻¹) (b) Org-P (µg P g-soil⁻¹) and (c) TP (µg P g-soil⁻¹).
Across all sites, no differences in the TP leached from soils were detected between treatments (p = 0.92) (Fig. 13), however. The organic-P, defined as the difference between TP and SRP, released is greater from soils leached after the T₂ treatment compared to the C treatment. Concentrations of TP released from the individual soil sites and treatments are shown in Table 6. The organic-P (the differences between SRP and TP) is represented in Fig. 10.

**Fig.10** Temperature experiment results by site A) Locations of all four sites where soils were collected for the temperature experiment (Bear River, Wasatch, West Uinta, East Uinta) B) Temperature treatments used for the experiment: Historical (C) represents average growing season temperatures from 2001-2005, recent (T1) represents average growing season temperatures from 2013-2017, and the projected (T2) temperatures represent temperatures 2 °C higher than T1 temperatures. The x-axis indicates the week of the year with the day represented in the experiment underneath in parentheses. C) Results from the temperature experiment showing the average amount of phosphorus leached (μg P g soil) following treatment treatments (C, T1, T2) for each of the 4 soil sampling sites (bottom) SRP, Org-P as a percentage of TP (top) Letters indicate significant differences between treatments.
The percent SRP and Org-P of the TP for each treatment are shown in Fig. 13c. Across all sites, the average percent SRP was higher in the T2 treatment at 22.3% compared to the C treatment at 5.7%, (p = 0.005, t-test). Inversely, the % Org-P was greater in the C treatment at 94.3% compared to the T2 treatment at 77.7% (p = 0.005).
DISCUSSION

The recent findings of nationwide increases in P concentrations in remote waterbodies, identified as “minimally disturbed,” lead us to investigate anomalous mechanisms for P inputs into water bodies. Our observations that remote waterbodies tend to be in high-elevation, mountainous regions prompted several hypotheses related to the unique climate characteristics of these environments, including high rates of precipitation and extreme seasonal temperature variability. Using the Uinta Mountains in northeastern Utah as a case study, we tested the feasibility of three potential mechanisms for increasing P concentrations in remote high-elevation catchments. We tested whether 1) dust deposited in the Uintas contains bioavailable-P, 2) increasing soil pH increases leaching of SRP, and 3) increases in soil temperature can stimulate the availability of SRP in catchment soils.

From our analyses, we found that 1) dust deposited in the Uintas contains considerable amounts of bioavailable-P, 2) experimental increases in soil pH increased the amount of SRP leached from soils, but there is no evidence in the Uinta mountains for significant increases in soil pH over the last several decades, and 3) experimental treatments for elevated soil temperatures suggest a causal relationship between temperature/extended growing seasons and elevated SRP release. Based on these findings, we suggest the most parsimonious explanation for increasing P in lakes and streams in the Uinta mountains are atmospheric dust and increasing temperatures with climate change. While increases in soil pH have the potential to drive increased inputs of P, there is no evidence for significant fluctuations in soil pH in the Uintas over the last two decades.
We suggest all three mechanisms have the potential to drive increases in aquatic P concentrations within mountain watersheds, but the relative importance of each of these mechanisms will most likely depend on the distance to nearby dust sources, catchment characteristics, and the history of acid deposition with a given catchment. The relative contribution of each mechanism across mountain systems in the Western US requires further investigation. In the following paragraphs, I will describe our findings and how each of these factors (distance to dust source, catchment characteristics, acidification history) may influence the three mechanisms evaluated in this study within other mountain watersheds.

**Atmospheric Dust**

In the Uinta mountains, dust deposition rates range from 3.6 to 6.6, with a TP flux of 9.4 mg P m$^{-2}$ yr$^{-1}$, and bioavailable P flux of 4.6 mg P m$^{-2}$ yr$^{-1}$. This bioavailable P flux represents a significant source of external P to the catchment, comparable to dust-P inputs in other mountain catchments in the Western US. Despite these current estimates, without long-term monitoring, the actual dust inputs of P to these ecosystems over the past several decades are unclear, but several lines of evidence suggest dust-P inputs in the Uintas have increased since the early 2000s. There is a regional trend of increases in dust deposition in the southwestern US (Brahney et al. 2013) and the continued increase in the deposition of dust proxies through this time period (see supplementary). The Uintas receives dust from several critical dust sources in the western US (Aarons et al. 2017; Munroe et al. 2019), which are sensitive to shifts in climate, vegetation and land use, and correlations between historical changes in land use and anthropogenic activity with
increases in lake production suggest Uinta alpine and subalpine lakes have responded to recent shifts in atmospheric chemistry (Hundey et al. 2014).

Several studies suggest dust is increasing in the Southwest US, and alpine catchments closest to local and regional dust sources are likely to be the most affected by nutrient additions from dust. Rates of dust deposition tend to decrease with increasing distance from the dust sources. For example, in the Wind River mountains in WY, the highest fluxes of dust occur in the SW region of the mountain range, closest to the Green River basin (Brahney et al. 2014). In the Uintas, we presume dust gradients are not as distinct because there are multiple dominant dust sources along the E-W transect of the mountain range contributing to alpine deposition, including the playas deserts in the Great Basin (W-NW of the Uintas) and arid basins in the Colorado Plateau (S-SW of the Uintas). This theory is supported by similar isotopic fingerprints between Uinta dust and sediment from multiple different dust source regions, as well as both SW and NW predominant wind directions (Munroe 2014; Munroe et al. 2019).

Other important factors controlling the flux and composition of dust include precipitation rates, wind directions, and atmospheric emissions from dust sources and anthropogenic activity (Lawrence and Neff 2009, Brahney et al. 2020), which may also explain differences in dust deposition between sites in the Western US. In this study, dust collected in the West Uinta site appears to have a greater portion of recalcitrant P fractions, indicating greater inputs of mineral-bound P in the West compared to the East Uintas. While these differences were not statistically significant, greater mineral dust inputs in the West Uintas is possible, given the proximity to playa dust sources in Western Utah (Dastrup et al. 2018), as well as greater rates of precipitation in the West
compared to the East region of the Uintas (MacDonald and Tingstad 2007). The presence of an active phosphate mine just east of the Uintas, may also explain the similar mineral-P contributions in the East, despite the greater distance from these western dust sources.

However, we might also expect to see a greater portion of bioavailable P in the West Uintas compared to the East as well, given the densely populated regions along the western Wasatch front. Previous work suggests anthropogenic P sources are more bioavailable (Anderson et al. 2010), and atmospheric emissions associated with burning fossil fuels may acidify dust during transport to the depositional environments, enhancing the solubility of mineral-P compounds (Gao et al. 2019). Yet, we observed relatively similar bioavailable P concentrations in the dust samples, although measurements had a significant amount of variability.

The sampling method used for summer deposition may overestimate the amount of bioavailable P fractions in dust due to the post-depositional chemical and biological transformations prior to dust collection. However, it is important to note that these conditions in the samplers are experienced by dust in the natural environment. For instance, in the summer dust, elevated NaOH-P and Bicarb-P compared to winter deposition may be explained by greater dissolution of dust in the samplers during the summer months from acidic precipitation and higher temperatures, followed by resorption by metal oxide minerals under low pH condition. When samples are dried and then rewetted, P can be released from microbial biomass, organic colloids dissolve, and inorganic P can become absorbed or diffused on mineral surfaces. These alterations tend to favor increases in salt-extractable, or loosely-sorbed fractions. (Condron and Newman 2011). In dust on snow samples, similar leaching and chemical alterations from the snow
may occur, but to a smaller degree. Limitations for the sequential extraction method in defining different bioavailable pools are discussed elsewhere, but to summarize, these extraction methods do not differentiate between specific P compounds, and there is potential residue and neutralization between extraction steps (Condron and Newman 2011). Furthermore, a limitation of using a unique sequential extraction method is the difficulty in making comparisons between different studies. For instance, SEDEX defines different fractions of P that do not correlate with ones used in our method (Levy and Schlesinger 1999).

Therefore, we also note that the flux and composition of dust seems to vary by season, an important consideration when estimating dust and P deposition rates. Dust-on-snow (representing winter and early spring) measurements may underestimate the bioavailable flux of P from dust. In the Uinta dust, the recalcitrant portion of P was similar between the winter and summer dust, but as previously noted, the summer dust had a greater fraction of bioavailable-P. While some of this elevated bioavailable P in the summer dust may be due to chemical transformations, the greater overall P enrichment in summer dust suggests there is more organic material in summer dust, also considering the greater NaOH-P fraction in summer deposition. Although NaOH-P also includes P bound to metal oxides. Differences in P fractions between summer and winter dust can be explained by seasonal shifts in dominant dust sources with changing wind directions and rates of dust emissions from major sources. In the Uintas, the prevailing wind direction shift from primarily NW in the fall/winter to primarily S in the spring/summer (Munroe et al. 2019). To the W and NW of the Uintas are dry playa sources in the southwest US, which are shown to have relatively consistent dust fluxes between seasons, but sites near
alluvial dust sources, such as those in the Colorado Plateau, had higher fluxes of dust in the summer-fall compared to the winter-spring (Reheis and Urban 2011), which is consistent with seasonal shifts in dust deposition observed in the Colorado front range (Heindel et al. 2020). Also, forest fires may create temporary dust sources that contribute bioavailable P to the atmosphere during the summer months (Vicars et al. 2010).

Altogether, as suggested by Munroe et al. (2019), future studies should also include summer deposition in estimating atmospheric P fluxes, especially as growing seasons become longer with climate change, which has already been linked to higher dust emissions in March and April throughout the southwestern US (Hand et al. 2016).

**Soil pH**

We found that acidification reduced the amount of P-released from Uinta soils, while the opposite-alkalinization-, increased the amount of P released. Our results confirm that the extent of P release and binding depends on the site characteristics. The different responses of the Eastern and Western Uinta soils to changes in pH reflect the variability of soil responses that occur even within a mountain range with relatively uniform bedrock. Soils collected from the alpine zone of the Uinta, with rocky catchments and relatively little vegetation released less SRP as the soils were alkalinized than the two lower elevation soils. Smaller fluxes of SRP from the alpine catchment compared to the subalpine catchments is consistent with decreasing inorganic P along elevational gradients (Sundqvist et al. 2014; Yang et al. 2016). It was also clear during the experiments that the different soil types responded differently to the additions of acids and bases, which is likely related to organic matter and clay content. Soils with high percentages of organic matter or clay typically have high buffering capacities, requiring
more H+ or OH- to alter the pH. Though we did not quantify the amount of acid or base required to change the pH of the soil, it may be important for future studies to measure the acid-neutralizing capacity to predict how sensitive the soils are to acidity.

Based on our assessment of historical soil pH values, it appears that the pH of the Uinta soils has not changed significantly over the last 20 years. However, if an increase in 0.1 pH occurs, our model predicts an increase in SRP ranging from 0.6-27 mg P m\(^{-2}\) in Uinta watersheds, which suggests that elevated soil pH may be an important mechanism within other catchments in the Western US experiencing acid recovery.

In most Western high elevation sites, historic concentrations of acidic ions in precipitation were not recorded as extensively as the populated industrial areas of the Eastern US. The location of National Atmospheric Deposition Program (NADP) sites in Northeastern Utah and Western Colorado are outside of the Uintas and provide only a broad evaluation of trends in atmospheric deposition across the region. However, NADP data show acid recovery has occurred within the last 20 years in the Colorado Front Range. The NADP site at Niwot Saddle, CO (CO-02) shows that atmospheric deposition of NO\(_3^-\) and SO\(_4^{2-}\) reached \(\sim27\) kg ha\(^{-1}\) and \(\sim15\) kg ha\(^{-1}\) around the year 2000 and has since declined, with deposition rates dropping to \(\sim10\) kg ha\(^{-1}\) and \(\sim4\) kg ha\(^{-1}\) in 2018 (Fig. 11) (Moser et al. 2019), which could in part explain recent increases in the trophic status of these lakes.

Given this evidence for atmospheric pH recovery in Colorado mountain regions, mountain catchments in these regions may exhibit elevated P fluxes, depending on how sensitive soils are to changes in pH, as well as other characteristics contributing to P retention in the catchment. Soil parent materials, biota, climate, and topography all
influence P pools and transformations, and shallow soil depth leads to less overall P retention in alpine catchments compared to lower elevation catchments. There is evidence for phosphorus limitation in high elevation soils of the Colorado front range (Bueno de Mesquita et al. 2020), thus increases in P may stimulate production in soils and increase P retention.

In this study, differences in soil responses between broad vegetation categories were unclear, and future research should focus on alternative soil categories using soil classifications reflecting landscape positions associated with different P translation and transformation processes (Sommer and Schlichting 1997; Homyak et al. 2014).

Soil Temperature Experiment

For all soils, except those collected from the East Uintas, the highest concentrations of SRP were leached from the T2 temperature treatment, representing the projected increases in temperature for high-elevation sites in the Uinta Mountains. These effects were more pronounced in the Wasatch and Bear River sites, indicating the composition of the soil and parent material are a significant factor in determining how sensitive the P-pools are to changes in temperature.

In the Uinta soils, it is possible that soils from the T2 treatment had lower TP/SRP from elevated microbial activity and greater uptake of P. The concentration of SRP in the soil-water solution represents the differences between the processes of immobilization and mineralization in the soils. Greater mineralization and less immobilization results in greater ambient SRP concentrations while greater microbial immobilization than mineralization results in less SRP and greater organic P concentrations. Previous work indicates the effects of warming on mineralization and immobilization processes are site
specific (Brooks et al. 1998), and differences in characteristics such as organic matter quality, moisture, and soil composition also influence these processes (Nadelhoffer et al. 1991).

Because this was an incubation experiment aiming to simulate natural conditions over a condensed growing season, we had to make trade-offs that limit the interpretation of our results. Notably, soil moisture may also be an important factor influencing P cycling that we did not test in this study. We began the incubation treatments with soils at approximately 50% saturation, but we allowed gas exchange through small holes in the plastic covering of the soil core. It seems likely that soils in the T2 temperature treatment experienced more water loss than T1 and C treatments. Considering water loss has been shown to inhibit microbial activity in warming soils (Allison and Treseder 2008), it is possible soil moisture may be a confounding factor in this study.

Increasing temperatures and extended growing seasons are occurring with climate change in alpine sites, and the effects of these increasing temperatures on the ecosystems are not well understood (Elser et al. 2020). In addition to the increases in SRP observed in this study, warming may enhance the P fluxes in alpine lakes by altering other biogeochemical processes in the catchment. For instance, more of the catchment will be exposed to processes of erosion as the duration and extent of snowpack declines (Rogora et al. 2003; Crawford et al. 2019; Kopáček et al. 2019). Changes in snowpack timing and duration has also been shown to influence the temperature and microbial activity in underlying soils, leading to changes in the storage and fluxes of NO3− from soils during spring snowmelt (Brooks et al. 1998). Furthermore, drying and rewetting of soils, which is predicted to occur with more frequency with climate change, has been found to
suppress microbial activity and increase the amount of water-soluble P in soils (Turner and Haygarth 2001; Allison and Treseder 2008).
CONCLUSION

The widespread increase in phosphorus concentrations in mountain waters and streams of the United States has stymied researchers and managers alike because traditional catchment sources of phosphorus, urban and agricultural runoff, are unlikely drivers. Speculation into the potential causes include broad-scale phenomena and have ranged from increased dust fluxes, increased precipitation and erosion, as well as climate-driven changes in weathering. Here, we tested two hypotheses put forth by Stoddard et al. 2019 including the widespread dust mediated transfer of P to lake systems and the alleviation of acid deposition. In addition, since mountain environments are warming relatively quickly, we examined whether warming mountain soils could increase the flux of phosphorus to receiving water bodies. Our results indicate the potential for all three mechanisms to play a role in mountain water eutrophication. However, the mechanism that dominates in any given region will vary with 1) parent geology and soil composition, 2) proximity to dust sources and dust composition, and 3) trends in acid deposition. We conclude that despite the widespread increases in TP across isolated high-elevation water bodies in the Western US, a singular driver for these increases is unlikely; mechanisms for P increases in mountain sites require investigation at the local and regional scale.
REFERENCES


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Fig. A-1 Soil pH values measured from soils grouped by site (WL, TL, HL) and vegetation type (Pine, Grass/shrub) Error bars represent the upper and lower standard error.

Fig. A-2 Comparison of percent loss-on-ignition (%LOI values) between the East Uinta, West Uinta, and Wasatch site. LOI is a proxy for % organic matter. The Wasatch soils had lowest %LOI compared to the other site, indicating less organic matter in those soils.
Fig. A-3 Log Transformation of y-axis (SRP ug/g soil) for linear model to meet the assumptions of ANOVA. (a) Data displayed is the SRP concentration of leachate adjusted to the soil mass (ug SRP / g soil) measured after adjusting to the pH to the 7 different pH values (n=9) Data displayed using linear x & y axis before transformation and (b) Data displayed with log transformed y-axis log [SRP ug/g soil] (c-d) Diagnostic plots for model to evaluate whether the assumptions of ANOVA are met after log transformation of y-axis.
Table A-1 Results from pH experiment SRP released per gram of soil for each pH value. TL is the alpine catchment in the East and HL is the subalpine catchment in the East.

<table>
<thead>
<tr>
<th>pH</th>
<th>TL (East) Pine (ug P/g soil)</th>
<th>Shrub/ grass (ug P/g soil)</th>
<th>HL (East) Pine (ug P/g soil)</th>
<th>Shrub/ grass (ug P/g soil)</th>
<th>WL (West) Pine (ug P/g soil)</th>
<th>Shrub/ grass (ug P/g soil)</th>
</tr>
</thead>
<tbody>
<tr>
<td>4</td>
<td>0.1 ±0.02</td>
<td>0.01 ±0.03</td>
<td>0.2 ±0.50</td>
<td>0.02 ±0.17</td>
<td>0.3 ±0.17</td>
<td>0.0 ±0.03</td>
</tr>
<tr>
<td>4.5</td>
<td>0.2 ±0.01</td>
<td>0.1 ±0.04</td>
<td>0.1 ±0.06</td>
<td>1.4 ±0.84</td>
<td>0.1 ±0.12</td>
<td>0.1 ±0.28</td>
</tr>
<tr>
<td>5</td>
<td>0.4 ±0.00</td>
<td>0.2 ±0.03</td>
<td>0.6 ±0.24</td>
<td>6.3 ±1.13</td>
<td>0.2 ±0.22</td>
<td>0.2 ±0.68</td>
</tr>
<tr>
<td>5.5</td>
<td>0.6 ±0.07</td>
<td>0.3 ±0.02</td>
<td>2.0 ±0.47</td>
<td>7.3 ±1.52</td>
<td>0.3 ±0.15</td>
<td>0.7 ±1.70</td>
</tr>
<tr>
<td>6</td>
<td>0.9 ±0.09</td>
<td>0.2 ±0.06</td>
<td>5.6 ±0.80</td>
<td>10.3 ±2.47</td>
<td>2.0 ±0.68</td>
<td>1.4 ±0.97</td>
</tr>
<tr>
<td>6.5</td>
<td>1.8 ±0.28</td>
<td>0.4 ±0.05</td>
<td>6.6 ±1.16</td>
<td>10.9 ±0.58</td>
<td>4.9 ±0.41</td>
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<tr>
<td>7</td>
<td>2.8 ±0.92</td>
<td>1.1 ±0.09</td>
<td>8.2 ±0.23</td>
<td>15.4 ±3.23</td>
<td>9.6 ±1.18</td>
<td>1.8 ±0.21</td>
</tr>
</tbody>
</table>

Fig. A-4 Extrapolated relationship between increase in pH and increases in the P released from soils, showing how the magnitude of the change depends on the starting pH of the soil. Dashed line (5.1 pH) represents the average initial soil pH for all soils measured.
Fig. A-5  Soil Temperature experiment treatments used in study

Table A-2  Average weekly air temperatures collected by SNOTEL sensors in alpine and subalpine catchments in the Uinta Mountains

<table>
<thead>
<tr>
<th>Experiment Day</th>
<th>Week of Year</th>
<th>C</th>
<th>T1</th>
<th>T2</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>Average T (°C) Day</td>
<td>Night T (°C) Day</td>
<td>Night T (°C) Day</td>
</tr>
<tr>
<td>1</td>
<td>21</td>
<td>-</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>2</td>
<td>22</td>
<td>6.2</td>
<td>10.5</td>
<td>0.2</td>
</tr>
<tr>
<td>3</td>
<td>23</td>
<td>7.0</td>
<td>11.1</td>
<td>1.4</td>
</tr>
<tr>
<td>4</td>
<td>24</td>
<td>8.3</td>
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<td>25</td>
<td>9.4</td>
<td>12.5</td>
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<td>8.8</td>
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<td>9</td>
<td>29</td>
<td>9.8</td>
<td>13</td>
<td>5.3</td>
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<td>9.6</td>
<td>12.5</td>
<td>5.4</td>
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<td>8.9</td>
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<td>8.4</td>
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<td>13</td>
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<td>11</td>
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<tr>
<td>14</td>
<td>34</td>
<td>-</td>
<td>-</td>
<td>-</td>
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</tbody>
</table>
Table A-3 Pairwise Mann-Whitney tests for temperature data by site. Mean represents the mean difference in SRP released from different temperature treatments (C, T1, T2)

<table>
<thead>
<tr>
<th>West Uinta</th>
<th>Bear River</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Pairwise Mann-Whitney tests</strong></td>
<td><strong>Pairwise Mann-Whitney tests</strong></td>
</tr>
<tr>
<td><strong>group</strong></td>
<td><strong>group</strong></td>
</tr>
<tr>
<td>1</td>
<td>2</td>
</tr>
<tr>
<td>C</td>
<td>T1</td>
</tr>
<tr>
<td>C</td>
<td>T2</td>
</tr>
<tr>
<td>T1</td>
<td>T2</td>
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</tbody>
</table>

<table>
<thead>
<tr>
<th>East Uinta</th>
<th>Wasatch</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Pairwise Mann-Whitney tests</strong></td>
<td><strong>Pairwise Mann-Whitney tests</strong></td>
</tr>
<tr>
<td><strong>group</strong></td>
<td><strong>group</strong></td>
</tr>
<tr>
<td>1</td>
<td>2</td>
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<td>C</td>
<td>T2</td>
</tr>
<tr>
<td>T1</td>
<td>T2</td>
</tr>
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</table>
Fig. A-6  Oxalate extracted Al, Fe, and Mn from soils
Fig. A-7  Soluble reactive phosphorus released after different temperature incubation treatments for different vegetation sites. Colors represent different vegetation types P = pine soils, S = grass/shrub soils. (a) Tony Grove (TG) soils (b) Willard Peak (WP) soils, (c) West Uintas - Wall Lake (W) soils (d) East Uintas - Chepeta Basin.
**Fig. A-8** Comparison of Soil pH measurements of soils from the 4 different sites used in the temperature experiment
Fig. A-9  Soil temperature trends in the Uinta mountains 2001-2018. Black dots represents sites where data were not available for the time period.
Fig. S-1  NADP measurements of nitrate and sulfate deposition in kg/ha at site CO-02.