

**Biogeochemical
response of alpine
lakes to changes in
dust deposition**

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Biogeochemical response of alpine lakes to recent changes in dust deposition

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Abstract

The deposition of dust has recently increased significantly over some regions of the western US. Here we explore how changes in dust deposition have affected the biogeochemistry of two alpine watersheds in Colorado, US. We first reconstruct recent changes in the mass accumulation rate of sediments and then we use isotopic measurements in conjunction with a Bayesian mixing model to infer that approximately 95% of the inorganic fraction of lake sediments is derived from dust. Elemental analyses of modern dust indicate that dust is enriched in Ca, Cr, Cu, Mg, Ni, and in one watershed, Fe and P relative to bedrock. The increase in dust deposition combined with its enrichment in certain elements has altered the biogeochemistry of these systems. Both lakes showed an increase in primary productivity as evidenced by a decrease in carbon isotopic discrimination; however, the cause of increased primary productivity varies due to differences in watershed characteristic. The lake in the larger watershed experienced greater atmospheric N loading and less P loading from the bedrock leading to a greater N:P flux ratio. In contrast, the lake in the smaller watershed experienced less atmospheric N loading and greater P loading from the bedrock, leading to a reduced N:P flux ratio. As a result, primary productivity was more constrained by N availability in the smaller watershed. N-limited primary productivity in the smaller watershed was partly ameliorated by an increase in nitrogen fixation as indicated by reduced nitrogen isotopic values in more contemporary sediments. This study illustrates that alpine watersheds are excellent integrators of changes in atmospheric deposition, but that the biogeochemical response of these watersheds may be mediated by their physical (i.e. watershed area) and chemical (i.e. underlying geology) properties.

1 Introduction

Nutrients are often transported between ecosystems, leading to the depletion of nutrients from source ecosystems and the enrichment of nutrients in sink ecosystems.

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Thus the composition and productivity of sink ecosystems may be dependent on the magnitude and composition of the so-called nutrient subsidies from source ecosystems (Polis et al., 1997). Atmospheric aerosols can be an important vector of nutrients from source ecosystems to sink ecosystems. This is especially true for arid regions where dust emissions can result in substantial nutrient subsidies to sink ecosystems in the form of Fe, P, and N deposition (Duce and Tindale, 1991; Neff et al., 2008; Okin et al., 2004). Recent evidence of increased dust emissions due to extensive grazing in the Western US suggests an increase in nutrient loading to nearby alpine watersheds (Neff et al., 2008). At the same time rapid urbanization of the Western US has lead to a massive and sudden increase in the amount of dry and wet deposition of N (Fenn et al., 2003). Thus changes in the intensity and patterns of human land use have greatly altered the biogeochemical cycles of N and P in the Western US, with wide-ranging impacts on adjacent ecosystems.

A large amount of research has focused on the ecosystem impacts of increased N deposition as a result of fossil fuel emissions by humans (Likens et al., 1996; Vitousek et al., 1997). It has been estimated that total N deposition in the form of NO_x and NH₃ has tripled since 1860 and is expected to double again by 2050 (Galloway et al., 2004). These increases in N-deposition have lead to increases in the %N of soils, decreases in the C:N of biomass, and an increase in NO₃ of surface waters in ecosystems adjacent to urban areas with increased N emissions (Baron et al., 2000; Fenn et al., 2003). However, human activity may have also resulted in a 30–50% increase in total P deposition in perturbed regions of North America, Asia, and Africa (Mahowald et al., 2008). Therefore, across these regions differential changes in the rates and amounts of N and P deposition may have altered the relative availability of these nutrients and ultimately biogeochemical processes occurring in these ecosystems.

The stoichiometric ratios of essential elements are an important constraint on the productivity and composition of ecosystems (Serner and Elser, 2002). Thus changes in the relative deposition and availability of N and P can have impacts on ecosystem structure and function. The relationship between atmospheric N deposition and nutrient

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limitation has been verified for a large number of Northern Hemisphere lakes, where high levels of N deposition result in greater N availability ultimately leading to P-limited primary productivity (Elser et al., 2009). However, much less research has focused on the extent to which changes in P deposition have altered ecosystem processes. It has been shown that increased human land-use, particularly grazing in the Southwestern US, resulted in a 400 to 500% increase in dust emissions approximately 100 years before present, resulting in increased P deposition in nearby alpine watersheds (Neff et al. 2008). Therefore shifts in human land-use towards more industrial practices in the Western US should result in an increase in the relative deposition of N:P leading to increased N availability in ecosystems. Such a temporal shift in the relative deposition of N:P could potentially have an impact on biogeochemical cycles and productivity in alpine ecosystems. However physical and chemical differences in watershed properties may dampen or amplify the impact of changes in N and P deposition on alpine ecosystems. For instance, lakes located in watersheds with a greater catchment to surface area ratio may be more susceptible to nutrient loading from the atmosphere and lakes located in watersheds with little or no P present in their bedrock may show a greater response to increased atmospheric P-loading.

Here we build upon previous observations that an increase in human activity has led to an increase in dust emissions in the Western US (Neff et al., 2008). In particular, we investigate the biogeochemical consequences of changes in dust deposition in alpine ecosystems adjacent to regions of known dust emission. We investigate changes in the biogeochemical response of two watersheds located in the San Juan Mountains (SJM) of Southern Colorado by analyzing lake sediment cores from their catchments. First, we establish recent changes in the physical flux of dust to these systems by estimating mass accumulation rates based on radionuclide dates and density estimates. We then apply geochemical techniques to lake sediments, atmospheric dust, and watershed bedrock to partition sediment fluxes between local endogenous sources (i.e. bedrock) and regional exogenous sources (i.e. dust). Lastly, we investigate how these changes in the physical flux and chemical composition of dust has impacted the productivity and

species composition in these systems. Although both of these watersheds experienced similar and significant increases in dust deposition, the biogeochemical response of these ecosystems was mediated by physical and chemical differences in watershed properties.

2 Study sites and methods

2.1 Study sites and sample collection

For this study we sampled two alpine watersheds in the SJM, which lie directly downwind of the Colorado Plateau, a known dust source region (Simonson, 1995) (Fig. 1). The underlying geology of the SJM has been shaped by Cretaceous plutonic activity, Tertiary volcanic activity, and glacial retreat approximately 12 000 year ago. Within the SJM we selected two watersheds- Porphyry and Senator Beck- based on their high elevations (approx. 3500 m), lack of upslope soils and vegetation, and their volcanic bedrock that is geochemically distinct from aeolian dust (Lawrence et al., 2010; Neff et al., 2008). At each site, sediment cores were extracted in Jul 2007 from small undisturbed tarns (Surface Area < 1000 m² and Depth < 1 m) using a Universal Corer. Sediment cores were transported on ice to the laboratory where they were extruded at a high resolution (0.5 to 1.0 cm) for geochemical analyses and ²¹⁰Pb dating, as well as plant macrofossils for ¹⁴C dating. A diverse range of bedrock types was sampled in each watershed for geochemical analyses. For Senator Beck tarn, we collected 9 bedrock samples and for Porphyry tarn we collected 8 bedrock samples. The outer weathering rind of all bedrock samples was first removed using a rock saw and then samples were pulverized using a shatterbox to yield homogenized samples for our geochemical analyses. Lastly, discrete and widespread dust deposition events were identified and collected from the snowpack within 2 to 3 days of deposition (Supplement: Fig. 1). Snowpack samples were melted, evaporated and then freeze dried prior to geochemical analysis, according to Lawrence et al. (2010).

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2.2 Dating of lake sediments

Surface sediment samples were dated using ^{210}Pb and sediments at depth were dated using ^{14}C . ^{210}Pb is continuously produced from the decay of ^{222}Rn gas in the atmosphere and because it has a half-life of ~ 22 years it is suitable for dating sediments up to ~ 150 years old. Bulk surface samples were digested and spiked with ^{210}Po and decay rates were estimated using an alpha spectrometer, according to the method described by DeMaster (1985). Unsupported ^{210}Pb activity levels were estimated for 12 samples per sediment core and sediment accumulation rates were calculated using the constant rate of supply model (Appleby and Oldfield, 1983). For dating older sediments, plant macrofossils were identified and removed from the sediments. These samples were rinsed, sonicated, and then freeze dried prior to analysis for ^{14}C content using the Keck-Carbon Cycle Accelerator Mass Spectrometer at UC Irvine. An oxalic acid standard of known ^{14}C content was analyzed to ensure the precision of dates (Neff et al., 2008). Because ^{14}C has a much longer half-life (5730 years) than ^{210}Pb (22 years) there is a notable gap in our dating techniques between our last reliable ^{210}Pb dates (147 ± 19 and 210 ± 33) and our first reliable ^{14}C dates (3138 ± 30 and 1163 ± 29). To infer changes in sediment accumulation during this interval of uncertainty piecewise spline functions were fit to the age-depth relationship and the first derivative of these spline functions was calculated as an estimate of changes in sedimentation rate (cm yr^{-1}) over time (see Sect. 2.4). These continuous functions of changes in sedimentation rate were then multiplied by sediment density (g cm^{-3}) in order to calculate changes in sediment accumulation rate ($\text{g cm}^{-2} \text{yr}^{-1}$). Changes in density for each sediment core were measured at the Limnological Research Center Core Facility at the University of Minnesota using a Geotek Multi-Sensor Core Logger, spurious density measurements at the sediment-water interface were not included in our mass accumulation calculations. Instrumental precision was between 0.01 and 0.05 g cm^{-3} and cores were logged with a depth resolution of 0.3 mm.

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2.3 Biogeochemical analyses

All sediment, dust, and bedrock samples were analyzed for elemental composition to identify the enrichment or depletion of elements in lake sediments relative to potential sources. Homogenized samples were treated with a series of digestions using HNO₃, HClO₄, and H₂O₂, followed by a microwave assisted digestion using HF and HCl to yield the mineral fraction of each sample. The mineral fraction of each sample was then analyzed for an array of elements using both Inductively Coupled Plasma Atomic Emission Spectroscopy (ICP-AES, ARL 3410+) and Inductively Coupled Plasma Mass Spectroscopy (ICP-MS, Perkin Elmer Elan DRC-E) at the Laboratory for Environmental Geoscience at CU Boulder. Two bedrock standards – Hawaiian Basalt (USGS BHVO-2) and Silver Plume Granodiorite (USGS GSP-2) – were also included in each extraction yielding an estimated precision of < 8% across all major elements. Elemental composition of the outer weathering rind and the inner unaltered bedrock were both analyzed and no significant difference in elemental composition was observed, suggesting no alteration of elemental composition as a result of weathering. We also analyzed 5 duplicate samples across both sediment cores and coefficients of variation ranged between 3% and 16% among all elements. Sediments were also analyzed for stable carbon ($\delta^{13}\text{C}$) and stable nitrogen ($\delta^{15}\text{N}$) isotopes. Sediment samples were treated with 10% HCl to remove the carbonate fraction and elemental carbon and nitrogen were measured using a Carlo Erba Elemental Analyzer and $\delta^{13}\text{C}$ and $\delta^{15}\text{N}$ were measured using a Finnigan MAT Delta Plus XL mass spectrometer at the Duke Environmental Isotope Laboratory. All samples were analyzed in combination with Acetanilide, cellulose, sucrose, and urea standards yielding an analytical error of $\sigma = 0.2\text{‰}$. All isotopic data are reported as δ values, defined as $\delta = (R/R_{\text{st}}) \times 1000$ where R is the ratio of isotopes in the sample and R_{st} is the ratio of isotopes in the standard. For carbon isotopes ($\delta^{13}\text{C}$) all values are reported with respect to the Vienna Pee Dee Belemnite (VPDB) standard ($R_{\text{st}} = 1.12372 \times 10^{-2}$) and for nitrogen isotopes ($\delta^{15}\text{N}$) all values are reported with respect to the atmospheric N₂ standard ($R_{\text{st}} = 3.68 \times 10^{-3}$).

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A subset of sediment, dust, and bedrock samples were also analyzed for isotopic composition of Sr, Sm, and, Nd to identify potential sources of allochthonous material to lake sediments. For Senator Beck sediments, two size classes were analyzed- the 37–60 μm size class which most closely resembles the particle size distribution of eolian dust (Neff et al., 2008) and the over 250 μm size class. Following the removal of ammonium-acetate-soluble material, samples were digested in a concentrated HF and HClO_4 solution and then evaporated to dryness. For Sr analysis, samples were added to 0.5 ml of 2 N HNO_3 in a Teflon column containing ~ 500 mg of Sr resin (Eichrom Technologies, Lisle, IL). These columns were then eluted with a total of 1.6 ml of 2 N HNO_3 and 3 ml of 7 N HNO_3 . Strontium samples were then collected in 1.5 ml of 0.5 N HNO_3 eluent for analysis. For Nd analysis, samples were eluted on a Teflon column with 1.6 ml of 2 N HNO_3 and then with 2 ml of 0.05 HNO_3 , which was collected in a clean Teflon vessel. These samples were evaporated to dryness, added to a calibrated glass-resin column in 50 μl of 0.25 N HCL, and Nd was collected with 0.25 N HCL. Lastly, $^{87}\text{Sr}/^{86}\text{Sr}$ and $^{143}\text{Nd}/^{144}\text{Nd}$ measurements were made using a Finnigan-MAT 261 thermal ionization mass spectrometer at the University of Colorado Radiogenic Isotope Laboratory. Analytical blanks included in the procedure averaged ~ 1 ng of Sr and ~ 100 pg of Nd. Thirty measurements of the Sr standard (SRM-987) during the study period yielded a mean $^{87}\text{Sr}/^{86}\text{Sr} = 0.71032 \pm 2$ and all reported $^{87}\text{Sr}/^{86}\text{Sr}$ values are corrected to the SRM-987 value of 0.71028. Thirty three measurements of the La Jolla Nd standard during the study period yielded a mean $^{143}\text{Nd}/^{144}\text{Nd} = 0.511838 \pm 8$ ($2\text{-}\sigma$ mean) and all reported $^{143}\text{Nd}/^{144}\text{Nd}$ values are normalized to $^{143}\text{Nd}/^{144}\text{Nd} = 0.7219$. Values of $^{147}\text{Sm}/^{144}\text{Nd}$ are calculated with a reproducible precision of 0.5% based on multiple measurements of an internal laboratory standard (Farmer et al., 2002).

Sediments were also examined for relative changes in diatom abundance and composition. Approximately 0.3–0.5 g of wet sediment from each sample was treated with 10% HCl to remove carbonates, followed by 30% hydrogen peroxide to remove organic material. The processed material was settled onto coverslips and mounted on slides with Naphrax[®]. A minimum of 300 diatom valves were counted for each sample

under oil immersion at a magnification of 1000X on an Olympus BX51 microscope with differential interference contrast. Diatom taxonomy was based on Krammer and Lange-Bertalot (1988) and Camburn and Charles (2000).

2.4 Statistical and geospatial analyses

The relationship between age, inferred from our radionuclide estimates, and depth of sediments was fit with a spline function and the derivative was calculated to estimate changes in sedimentation rate. The spline function was fit using the “smooth.spline” function (R 2007). Spline functions were fit using an intermediate smoothing parameter of 0.5 and weighted by the inverse of the error for each date inferred from our radionuclide measurements. Thus the more modern and precise ^{210}Pb dates were given more weight in the fitting process than the older and less precise ^{14}C dates.

For partitioning the allochthonous fluxes to our lake sediments we relied on the Bayesian mixing model developed by Parnell et al. (2010). This mixing model has advantages over conventional mixing models because it allows for multiple sources and multiple isotopes while providing posterior probability distributions rather than finite solutions for different sources. The model structure for inferring any number of sources (k), in this case bedrock and dust, contributing to an observation of isotopic (i) variability in a sample (j), such that the observed values (X_{ij}) in the sediment can be expressed as:

$$X_{ij} = \frac{\sum_{k=1}^K p_k q_{ki} s_{ki}}{\sum_{k=1}^K p_k q_{ki}} + \varepsilon_{ij}, \quad (1)$$

where p_k represents the proportions of k , contributing to X_{ij} , q_{ki} represents the concentration of isotope i in source k , s_{ki} represents the isotopic value of source k with a normal distribution of mean μ_{ki} and variance ω_{ki}^2 , and ε_{ij} represents residual

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inter-sample error not described by the model. We have specified the Dirichlet distribution for the prior of p_k , which is essentially the multivariate generalization of the Beta distribution (Parnell et al., 2010). Posterior distributions of p_k are then fit using a Markov chain Monte Carlo simulation. Multivariate models were evaluated based on optimizing coefficients of determination (R^2) and minimizing Akaike's information criteria (AIC). Principal components analyses were also performed on changes in elemental concentrations with age for each watershed (R 2007).

Morphometric properties of our study lakes and their watersheds were calculated using Arc-Map software (ArcGIS v. 9.3, ESRI, Redlands, CA). A handheld global positioning instrument was used to georeference both lakes. These georeferenced points were then overlaid onto orthophotos (Usda, 2010) in order to calculate lake surface area with the Arc-map measure tool. Interpolated 5 m digital elevation models (Usgs, 2010) were used to calculate the watershed area of each lake with the spatial analyst hydrology tools in Arc-map. This was done by first determining water flow direction for each watershed with the flow direction tool. Once the flow direction was established and verified, a layer containing lake pour points was developed using the watershed tool. The lake watershed area rasters were then converted to shape files and their surface areas were estimated using spatial statistic tools in Arc-map.

3 Results

3.1 Changes in the flux of dust to alpine catchments

Both Porphyry and Senator Beck Lakes have experienced considerable changes in the mass flux to sediments in the recent past. Dates inferred from ^{210}Pb in both sediment records indicate a much more rapid sedimentation rate in the last ~150 yr than during the previous Holocene interval (Fig. 2). In Porphyry, Holocene sedimentation rates inferred from ^{14}C were approximately 0.1 mm yr^{-1} , but increased to over 0.4 mm yr^{-1} at some point in the last 200 years (Fig. 2b). A similar increase in

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Senator Beck is observed, where sedimentation rates increased from approximately 0.06 mm yr^{-1} to over 0.2 mm yr^{-1} (Fig. 2f). Both lakes show an increase in sediment density with depth that one would expect as sediments become more compressed over time (Fig. 2c and g). Despite this increase in density with depth, both sediment records show a marked increase in contemporary mass accumulation rates compared to background Holocene mass accumulation rates (Fig. 2d and h). In Porphyry Lake mass accumulation rates increased from $\sim 100 \text{ g m}^{-2} \text{ yr}^{-1}$ to $\sim 500 \text{ g m}^{-2} \text{ yr}^{-1}$ (Fig. 2d) and in Senator Beck Lake mass accumulation rates increased from $\sim 100 \text{ g m}^{-2} \text{ yr}^{-1}$ to $\sim 400 \text{ g m}^{-2} \text{ yr}^{-1}$ (Fig. 2h). Although it is clear that these two lakes have experienced a 400 to 500% increase in mass accumulation over the last 200 years; it is unclear how much of this increase in mass accumulation can be attributed to atmospheric dust versus material derived from bedrock weathering in the watershed. Our previous qualitative analysis indicated that the inorganic fraction of sediments in these systems is primarily derived from regional dust sources and not from local bedrock weathering; however, we were unable to provide quantitative estimates of how much inorganic mass was derived from dust (Neff et al., 2008).

Based on the isotopic composition of inorganic material measured in dust, bedrock, and sediment samples, sediments are primarily derived from atmospheric dust and to a lesser extent from bedrock weathering (Table 1). Despite the fact that bedrock material in the SJM tends to have a higher concentration of Sr than material blown in as atmospheric dust, $^{87}\text{Sr}/^{86}\text{Sr}$ values of sediments are almost indistinguishable from $^{87}\text{Sr}/^{86}\text{Sr}$ values of dust. Although concentrations of Nd and Sm did not differ that much between sediments, dust, and bedrock, isotopic ratios of $^{143}\text{Nd}/^{144}\text{Nd}$ and $^{147}\text{Sm}/^{144}\text{Nd}$ differed considerably in these substrates. In fact, sediment values of $^{143}\text{Nd}/^{144}\text{Nd}$ and $^{147}\text{Sm}/^{144}\text{Nd}$ were much more similar to dust than bedrock, with dust values of $^{143}\text{Nd}/^{144}\text{Nd}$ and $^{147}\text{Sm}/^{144}\text{Nd}$ intermediate between sediment and bedrock values. These conservative isotopic tracers provide compelling evidence that sediments being deposited in these alpine watersheds largely reflect the geochemistry of dust being deposited in these alpine watersheds. This is verified by our mixing model

simulations that indicate 98% of the inorganic material accumulated in these alpine lake sediments is derived from atmospheric dust and 2% is derived from bedrock weathering (Fig. 3).

3.2 Biogeochemical response to changes in dust loading

5 Although increases in dust loading are observed in both Porphyry and Senator Beck watersheds, the biogeochemical response is quite different in these watersheds. Increases in organic carbon content in the modern sediments are evident in both lakes (Neff et al., 2008); however, the precise mechanisms driving these increases in organic carbon appear to be different in each lake. The organic carbon, organic nitrogen, and phosphorus content in sediments from Porphyry Lake are two to three
10 times greater than the organic C, organic N, and P content of sediments from Senator Beck (Fig. 4). Furthermore, increases in organic carbon in Porphyry (Table 3) are more responsive to increases in N concentration ($R^2 = 0.914$; AIC = 55.36) than to increases in P concentration ($R^2 = 0.701$; AIC = 118.89), with very little evidence of co-limitation by N and P ($R^2 = 0.915$; AIC = 55.35). However, there is a much stronger response in organic C to N when P content exceeds 0.1% in Porphyry sediments (Fig. 4c). Increases in organic C content are also highly correlated with N content in Senator Beck ($R^2 = 0.956$; AIC = -60.19), but correlated to a lesser degree with P content ($R^2 = 0.335$; AIC = 23.33). The relationship between organic C and organic N
20 as well as P is much stronger ($R^2 = 0.968$), as indicated by the reduced AIC (-68.51), suggesting possible co-limitation of primary productivity by N and P in Senator Beck. Lastly, there appears to be a strong response in organic C content to N availability even when P is in short supply in Senator Beck (Fig. 4f).

25 Stable isotopes in sediments provide independent evidence of increases in primary productivity in the two watersheds investigated, but suggest different sources of nitrogen fueling this increase in primary productivity. In both Porphyry and Senator Beck Lake sediments there was a clear increasing trend in $\delta^{13}\text{C}$ over the last 5000 years (Fig. 5). The stable isotopic composition of organic carbon is often used as a proxy of

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primary productivity in aquatic ecosystems, with increases in isotopic enrichment often used as an indicator of increased primary productivity (O'Reilly et al., 2003). However, the $\delta^{13}\text{C}$ of atmospheric CO_2 has also varied over the Holocene and more recently as a result of fossil fuel combustion (Francey et al., 1999), therefore we must correct for changes in $\delta^{13}\text{C}$ of source CO_2 to our alpine watersheds. Expressing our $\delta^{13}\text{C}$ values of organic carbon (δ_o) as discrimination against atmospheric CO_2 (δ_a), we obtain the discrimination equation $\Delta^{13}\text{C} = (\delta_a - \delta_o)/(1 + \delta_o)$ of Farquhar (1989). Using this approach we see a clear decrease in $\Delta^{13}\text{C}$ over the last 5000 years with a particularly strong decrease over the last 100 to 200 years in both watersheds (Fig. 5c and f). Thus, as rates of primary productivity increased and the pool of available dissolved inorganic carbon was reduced the degree to which algae discriminate against the heavier ^{13}C has diminished (Brenner et al., 1999). Although there is considerable variability in $\delta^{15}\text{N}$ in the organic fraction of sediments in Porphyry ($1.5 < \delta^{15}\text{N} < 4.0$), there is no clear trend over time in response to dust loading to Porphyry (Fig. 5a). In contrast, Senator Beck shows a clear decline in $\delta^{15}\text{N}$ over the last 5000 years with $\delta^{15}\text{N}$ values in contemporary sediments showing the greatest change and approaching the atmospheric value of 0.

An increase in fluxes of elements associated with dust is observed in the contemporary sediments from both watersheds. If we compare elemental concentrations in sediments to sediment dates based on our age model in a principal components analysis, we see that most of the variability observed over time can be explained by elements commonly associated with dust (Fig. 6). In Porphyry, most of the variance in elemental fluxes is explained by the first two principal components. Elements commonly associated with dust, such as Si, Na, Mn, K, Al and Fe contribute the most to the first principal component and other elements also associated with dust, such as P, Cu, and Ca contribute most to the second principal component in Porphyry (Fig. 6a). Elements such as P, Cu, Ca, Ni, and Sr are associated with contemporary sediments during the period of maximum accumulation rates due to dust inputs (Fig. 2d). In contrast, most of the variance in elemental concentrations observed in Senator Beck can be explained

by the first principle component (Fig. 6b). Elements that contributed most to this first principle component in Senator Beck were also base cations, such as Ca, Mg, Na, and K, but also metals often associated with dust, such as Cu, Al, Ti, and Fe. Increases in Ca, Cd, Cr, and Ni concentrations were associated with more contemporary sediments deposited during periods of maximum mass accumulation due to dust in Senator Beck (Fig. 2h).

Many of the elements that showed a relative increase in contemporary sediments based on our principle components analysis also tend to be enriched in dust samples collected from the snowpack of the SJM (Fig. 7). The elements that were consistently enriched in dust relative to bedrock in both watersheds were Ca, Cr, Cu, Mg, and Ni. There was an abundance of elements that were consistently depleted in dust relative to bedrock in both watersheds, but some of the notable elements were Al, K, and Mn. Lastly, there were several elements that showed inconsistent patterns of enrichment in dust with respect to the differing bedrock in the two watersheds (Fig. 7). Dust samples were enriched in Fe, Cd, P, and Sr compared to bedrock in Porphyry, but dust samples were relatively depleted in these particular elements when compared to the bedrock chemistry of Senator Beck. The differential enrichment of elemental dust loading in these two watersheds may have important implications for biogeochemical cycling within these two watersheds.

Changes in diatom assemblage provide independent evidence of major biological reorganization in response to increased dust loading to alpine watersheds of the SJM. Shifts in diatom species composition were observed in both cores. The most dominant species present in both sediment cores was *Staurosirella pinnata* (Ehrenberg) (Williams and Round, 1987), representing between 20 and 90% of the total diatom assemblage in any particular time interval (Fig. 7). Although *S. pinnata* is the most abundant diatom, it shows different trends in the two sediment cores. In Porphyry there is a gradual increasing trend in *S. pinnata* over the last 150 years, with almost complete dominance by this taxon over the last 50 years (Fig. 7a). In contrast, *S. pinnata* has remained dominant and fairly constant in Senator Beck over the last 500 years,

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with exception to the interval between 50 and 75 years ago when its relative abundance decreased by a factor of 2.5 (Fig. 7b). Both watersheds also show a different response in *Navicula* spp. In Porphyry, *Navicula* spp. peaked in abundance approximately 125 years ago and have since shown a gradual decline in abundance (Fig. 7a).

5 On the other hand, *Navicula* spp. in Senator Beck are less than 10% of the assemblage over the last 500 years, except for a peak at approximately 25% of the total diatom abundance roughly 75 years ago (Fig. 7b). The one diatom species that shows a coherent response in both watersheds is *Achnanthydium minutissimum* (Kützing) Czarnecki (Fig. 7), which is a fairly prevalent taxon in the SJM region (Sgro et al. 2007).
10 In both watersheds a gradual increase in the relative abundance of *A. minutissimum* has been observed with a peak in abundance between 50 and 75 years ago, followed by a decline and then a subsequent increase over the last 20 years. The coherent response in *A. minutissimum* and its temporal covariance with changes in elemental fluxes associated with dust suggest that this taxon may be sensitive to changes in the
15 magnitude of dust deposition and its chemical composition. There was also an apparent increase in *Encyonema minutum* (Hilse) D. G. Mann that appears to parallel the increasing trend in *A. minutissimum*; however, this similar response is only evident in Senator Beck (Fig. 7b).

4 Discussion

20 4.1 Changes in dust fluxes and their geochemical composition

Our previous work showed that mass accumulation rates in the SJM increased substantially over the past 200 years in response to an apparent increase in dust deposition (Neff et al., 2008). However, we were unable to provide quantitative estimates of how much of this increase in mass accumulation could be attributed to increases in dust
25 deposition. Here, we have demonstrated that dust is the major contributor to the inorganic fraction of lake sediments in the SJM. In fact, when we consider all of our isotopic

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tracers and their concentrations in our mixing model, we see that approximately 98% of the inorganic flux to these sediments is derived from material geochemically similar to modern dust and only 2% of the inorganic flux appears to be derived from bedrock in the watershed.

5 These estimates of the dust fraction in sediments greatly exceed estimates of dust contributions to soils in the SJM that are on the order of 10 to 40% (Lawrence, 2010). The disparity between these estimates suggests that these lakes are integrating a greater temporal span or a greater spatial extent of dust deposition than the soils. The estimated dust fraction in sediments is approximately 3 to 4 times that of soils,
10 which can be explained in part by the limited snow-free season in these alpine watersheds spanning approximately 4 months from June to September (Painter et al., 2007). Although much of the dust contributing to soils in these alpine watersheds is thought to be deposited during winter months (Lawrence et al., 2010), lakes are probably integrating dust deposition directly during summer months and indirectly through run-off during winter months. In fact, it has been shown that wet surfaces are 1.5 to 4 times more efficient at collecting atmospheric deposition of Ca, K, Na, and Cl than dry surfaces (Lewis, 1983), thus it is not surprising that the lakes investigated in this study would be more effective dust collectors than the surrounding dry substrates in their watersheds. Sedimentation rates are also highly variable within individual lakes,
15 with higher sedimentation rates often observed at greater depths. This phenomenon, known as “sediment focusing” (Davis, 1982), may also apply to the watershed scale where material deposited within the catchment is transported through avalanching or run-off. Such sediment focusing would lead to greater concentrations of dust within our lakes, especially at depth where our sediment cores were collected. Thus lakes
20 are probably integrating more of an atmospheric dust signal over a larger area during winter months and are probably more effective at capturing dust during summer months. Lastly, the relatively high proportion of dust in sediments suggests that alpine lakes are excellent recorders of changes in dust deposition and that there is very little geochemical influence from surrounding bedrock or soils in these watersheds.

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There is no appreciable change in the isotopic geochemistry of lake sediments with depth indicating that atmospheric dust fluxes have always been an important input to these alpine watersheds. For instance $^{87}\text{Sr}/^{86}\text{Sr}$ values in surface sediments (0.71814) were not that different from values at 6 cm sediment depth (0.71649) or values at 23 cm sediment depth (0.71699), none of which are significantly different from modern dust values (0.71632) (t-test; p -value = 0.06). However, $^{87}\text{Sr}/^{86}\text{Sr}$ sediment values were significantly different from bedrock (0.70797) (t-test; p -value = 0.00013). Therefore, assuming that the isotopic signature of regional dust sources in the Southwestern US has not changed appreciably, we must infer that dust deposition has always contributed to the mass flux in these alpine watersheds but that this dust flux has increased in recent years. Lastly, an increase in contemporary dust fluxes would also result in an increase in base cations and nutrients conveyed by dust with potential implications for the biogeochemistry of these alpine watersheds.

4.2 Implications for biogeochemistry

One of the most dominant paradigms in biogeochemistry is that primary productivity in lakes is primarily P-limited (Schindler, 1977). This paradigm is based on the strong relationship between P and chlorophyll concentrations in freshwater ecosystems (Vollenweider, 1971). However, more recent evidence from a wider array of lakes suggests that co-limitation of primary productivity by N and P is much more prevalent than previously thought (Lewis Jr. and Wurtsbaugh, 2008). A recent series of lake surveys and experiments revealed that P-limitation was much more prevalent in lakes subjected to higher levels of N deposition, whereas N-limitation was more prevalent in lakes subjected to lower levels of N deposition (Elser et al., 2009). Although both alpine watersheds in this study were subjected to an increase in atmospheric dust deposition, they show very different chemical and biological responses. Both lakes in this study show an increase in primary productivity in response to increased atmospheric dust loading; this is evidenced by the previously reported increases in organic C fluxes (Neff et al., 2008) and corroborated by the decrease in $\Delta^{13}\text{C}$ in more contemporary sediments

(Fig. 5). However, these lakes appear to respond very differently to the relative availability of N and P. The percent of organic C present in the sediments of both Porphyry and Senator Beck was highly correlated with the percent organic N (Fig. 4 and Table 2), indicating that primary productivity in these systems may ultimately be constrained by N availability. This is consistent with evidence from similar low N deposition lakes in Colorado, showing significantly diminished C:P and N:P ratios of suspended particulate matter (Elser et al., 2009). Although organic C percent in both of these lakes was not nearly as correlated with percent P (Fig. 4), there is evidence of co-limitation by both N and P in Senator Beck (Table 2), especially at intermediate P levels (Fig. 4f). The apparent difference in nutrient limitation in these two watersheds is due in part to the differences in elemental fluxes derived from dust versus bedrock in these two systems. Thus the physical and chemical properties of individual watersheds may help to explain the remaining variance in nutrient-primary productivity relationships often observed in freshwater ecosystems (Lewis Jr. and Wurtsbaugh, 2008).

Broad patterns of elemental enrichment by dust are observed relative to bedrock in both watersheds; however, there are some key distinctions between the watersheds. Several of the elements that were associated with contemporary sediment fluxes in our principal components analysis (Fig. 6) are also enriched in dust relative to local bedrock (Fig. 7). Elements such as Mg, Cu, and Ca are consistently enriched in dust and contribute heavily to the amount of variance explained in modern sediments. Mg and Ca are elements that are often associated with the finer soil fraction that is particularly susceptible to erosion as a result of grazing in the southwestern US (Neff et al., 2005). Thus, some of the elements enriched in dust and contemporary sediments may be derived from regions in the Western US that have been historically overgrazed. Some elements contained in dust, however, show differing patterns of enrichment in the two watersheds (Fig. 7). In particular, Fe, P, Cd, and Sr, are enriched in dust relative to the bedrock in Porphyry, but relatively depleted in dust relative to the bedrock in Senator Beck. The concentration of P in the bedrock of Senator Beck's watershed is approximately 4 × the concentration of P in the bedrock of Porphyry's watershed

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(Table 3). Furthermore, it is not so surprising that Fe and P show similar patterns in these two watersheds, as P and Fe cycles are closely coupled by the adsorption of orthophosphate to iron hydroxides under aerobic conditions typically found in small well-mixed alpine lakes, such as Porphyry and Senator Beck (Jensen et al., 1992). In fact, it appears that Fe and P are more closely associated in Senator Beck (Fig. 6b) suggesting that the P derived from bedrock may be more tightly bound to Fe and thus less available for primary productivity than the P derived from dust (Fig. 6). This has been corroborated by leaching experiments demonstrating that most of the P content in dust is associated with the easily remineralized organic fraction (Lawrence et al., 2010). Thus dust is consistently enriched in certain elements and other elements such as P that show variable enrichment may be.

Although differences in the chemical composition of underlying bedrock are obviously important in mediating the biogeochemical response of watersheds to changes in atmospheric deposition, in order to estimate changes in elemental fluxes we must also consider the differences in morphometric properties of these watersheds. The two lakes investigated in this study have very different morphometric properties with respect to their watersheds that have affected the elemental fluxes in these two watersheds. Porphyry has a much larger surface area (6920 m²) and also has a much larger watershed area (165 910 m²) and therefore integrates a larger atmospheric area than Senator Beck (110 m²), which has a much smaller watershed area (1420 m²). Porphyry also has a watershed to surface area ratio (24:1) that is nearly twice as big as that of Senator Beck (13:1), suggesting that Porphyry may be more sensitive to changes in the rate and composition of atmospheric deposition. Although both lakes experienced mass accumulation rates during the Holocene of approximately 100 g m⁻² yr⁻¹, mass accumulation rates in Porphyry increased by a factor 5 (~500 g m⁻² yr⁻¹) compared to mass accumulation rates in Senator Beck that increased by only a factor 4 (~400 g m⁻² yr⁻¹). Thus lakes with larger watershed areas and greater watershed to surface area ratios may be more impacted by changes in atmospheric deposition. However, mass accumulation rates of sediment may vary spatially and temporally even within a single lake

and therefore changes in mass accumulation rates should be interpreted with caution. In fact, the heterogeneous rate of sedimentation within lakes, often referred to as “sediment focusing” is a well recognized problem (Davis, 1982). Furthermore, as deep lake basins fill, their lake bottom area becomes reduced and thus even if the mass flux to the sediments remained constant there would be an apparent increase in mass accumulation rate. However, the lakes investigated in this study are fairly small and not very deep; thus we do not suspect that sediment focusing has lead to changes in the mass accumulation rates observed in these lakes. Knowledge of modern dust fluxes, weathering fluxes, and their respective chemical properties, as well as watershed dimensions allows us to calculate the mass balance of important elements, such as phosphorus and nitrogen, to lake sediments within the two watersheds (Table 3). Mass accumulation rates of dust in the SJM measured from the snowpack are approximately $10 \text{ g m}^{-2} \text{ yr}^{-1}$, which is roughly 3 times estimates of bedrock weathering ($3 \text{ g m}^{-2} \text{ yr}^{-1}$) for the SJM (Lawrence and citations within). Phosphorus concentrations in dust ($902 \mu\text{g g}^{-1}$) are relatively low compared to P concentrations in the bedrock of the Senator Beck Watershed ($1111 \mu\text{g g}^{-1}$) and relatively high compared to P concentrations in the bedrock of Porphyry ($288 \mu\text{g g}^{-1}$). Even though the concentration of P is much higher in the bedrock of Senator Beck, the smaller watershed to surface area ratio yields a mass flux of P from bedrock weathering ($47 \text{ mg m}^{-2} \text{ yr}^{-1}$) that is almost identical to the mass flux of P from bedrock weathering in Porphyry ($46 \text{ mg m}^{-2} \text{ yr}^{-1}$). Generally, P fluxes derived from atmospheric dust deposition are much higher than fluxes from bedrock weathering because the dust fluxes are greater. However, assuming that all atmospherically-derived P is ultimately transported to lake sediments, atmospherically-derived P fluxes in Senator Beck ($125 \text{ mg m}^{-2} \text{ yr}^{-1}$) are approximately half the atmospherically-derived P fluxes in Porphyry ($225 \text{ mg m}^{-2} \text{ yr}^{-1}$). Therefore the total P loading from weathering and atmospheric deposition based on our mass balance calculations are considerably higher in Porphyry ($271 \text{ mg m}^{-2} \text{ yr}^{-1}$) than in Senator Beck ($172 \text{ mg m}^{-2} \text{ yr}^{-1}$).

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The greater mass balance P flux estimates in Porphyry than Senator Beck based on modern direct measurements are consistent with greater inferred P fluxes in the sediments of Porphyry (Neff et al., 2008); however, sediment P fluxes inferred from both lakes are approximately double our P flux estimates based on mass balance, possibly
5 due to the greater fraction of autochthonous organic material in more contemporary sediments. Similar increases in P content of contemporary sediments have been observed for the nearby Uinta Mountains of Utah (Reynolds et al., 2010), perhaps indicating a more coherent regional change in the chemical composition of atmospheric deposition. However, increases in P content of sediments in the Utah lakes were attributed to the recent increase (~1960) in nearby P mining (Reynolds et al., 2010),
10 whereas P accumulation rates in the SJM began to increase much earlier (~1800) probably due an increase in the prevalence and intensity of grazing (Neff et al., 2008). Lastly, background P accumulation rates were approximately $100 \text{ mg m}^{-2} \text{ yr}^{-1}$ which is approximately double the P fluxes derived from bedrock weathering alone, suggesting that atmospherically-derived P loading has always been an important source of P to these alpine watersheds but that contemporary atmospherically-derived P loading has increased.

Similarly, we can estimate the mass balance of N in our two watersheds. Assuming that the underlying bedrock has very little N, almost all of the N loading to these watersheds is derived from the atmosphere. Based on our estimates of annual dust flux and the concentration of N in dust ($4400 \mu\text{g g}^{-1}$) and correcting for the different watershed to surface area ratios, we see that total N fluxes to these two watersheds are quite different (Table 3). The total N flux to Porphyry ($1099 \text{ mg m}^{-2} \text{ yr}^{-1}$) is nearly double the total N flux to Senator Beck ($611 \text{ mg m}^{-2} \text{ yr}^{-1}$). Although these N flux estimates are approximately 25% of the N flux estimates inferred from sediments, they are in agreement with the greater N flux inferred from the sediments of Porphyry. The greater inferred N fluxes from the sediments are probably the result of an increase in autochthonous organic material in the contemporary sediments and also the fact that we only effectively measured the dry N deposition captured by the snowpack in the
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winter. If we were to include wet N deposition our estimates of modern N flux would increase considerably.

Changes in diatom assemblage provide independent evidence of biological reorganization in response to increased dust loading to alpine watersheds of the SJM. Shifts in diatom assemblages were observed in both Porphyry and Senator Beck during the interval in which we suspect that dust deposition reached its maximum. In both Porphyry and Senator Beck there was a clear and coherent increase in *A. minutissimum* between 50 and 75 years ago. Although there are no rigorous transfer functions correlating particular diatom taxa with specific environmental parameters for the SJM, in low alkalinity lakes from other regions, the pH and TP optima for *A. minutissimum* and *S. pinnata* are quite similar, with pH optima between 7 and 8 and TP optima between 8 and 14 $\mu\text{g L}^{-1}$ (Camburn and Charles, 2000; Dixit et al., 1999). A regional diatom survey in the SJM also found that *A. minutissimum* was abundant at sites with elevated pH levels between 7.8 and 8.0 (Sgro et al., 2007). Such elevated pH levels would result from an influx of base cations associated with dust, but the optimal range of TP is hardly indicative of eutrophic conditions, suggesting that an increase in dust-derived P resulted in only a modest increase in primary productivity. It is also possible that the coherent response of *A. minutissimum* is the result of a common climatic driver leading to changes in lake level at this time. Although broad patterns of drought in the Western US are evident from 1930–1940 and from 1950–1960 (Cook et al., 2004), it is unclear whether these broad patterns were consistent at the local scale in the SJM and it is very difficult to correlate events in our low-resolution data with higher resolution climate data.

In summary, differences in the underlying bedrock chemistry and watershed morphometry have altered how these two watersheds have responded to similar increases in dust loading. Although Senator Beck has surprisingly high concentrations of P in the underlying bedrock, it has a relatively low watershed to surface area ratio. In contrast, Porphyry has a much lower P concentration in the underlying bedrock with a higher watershed to surface area ratio. Thus Senator Beck receives a greater fraction

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of endogenous P from the watershed but the overall P loading is greater for Porphyry (Table 3). Similarly the total N loading is much higher for Porphyry than Senator Beck because of its larger watershed to surface area ratio. Ultimately, the molar ratio of the N:P flux is higher in Porphyry (9.0) than in Senator Beck (7.9). Although both of these ratios are well below the Redfield ratio (16:1) indicative of N-limitation of primary productivity in both of these watersheds, the lower N:P ratio of fluxes to Senator Beck indicates that N-limitation should exert greater control on primary productivity in this system. In fact, this is consistent with the gradual depletion of $\delta^{15}\text{N}$ observed in the sediments of Senator Beck that is most likely due to N fixation. Although increases in N deposition due to fossil fuel combustion also may result in depleted $\delta^{15}\text{N}$ values (Wolfe et al., 2001), N deposition in the SJM is not particularly high and we would expect regional patterns in N deposition to be reflected in both sediment records. Furthermore, the onset in progressive depletion in $\delta^{15}\text{N}$ observed in Senator Beck is approximately 5000 years before present, which is well before widespread industrial processes in the western US. Based on a survey of Northern Hemisphere lakes (Elser et al., 2009) the onset of N-limitation of primary productivity should occur below an N:P ratio of atmospheric deposition of approximately 75. This threshold is considerably higher than the ratios estimated in our watersheds, which are classified as lakes of “low N deposition” according to Elser et al. (2009). However, these different values can be partially reconciled by the fact that we did not measure wet N deposition and thus our estimates of total N:P flux are probably biased low and in their survey Elser et al. (2009) did not consider P derived from underlying bedrock and thus their threshold values are probably biased high. Changes in lake chemistry in response to atmospheric deposition have been shown to vary based on bedrock chemistry in other alpine watersheds of Colorado (Mast et al., 2010). Despite the widespread reduction in sulfur emissions (Likens et al., 1996), SO_4 concentrations in some alpine lakes of Colorado have actually increased due to increased weathering of FeS_2 in their catchments (Mast et al., 2010). These studies highlight the importance of underlying geology in mediating the biogeochemical response of alpine ecosystems to global change. Regardless, we can

conclude that alpine lakes and specifically their diatom communities are sensitive to changes in the magnitude and chemical composition of atmospheric deposition and their degree of sensitivity may be determined in part by their physical and chemical properties.

5 Conclusions

Lakes are bellwethers of global change because they integrate physical, chemical, and biological processes occurring in their watersheds (Williamson et al., 2009). Alpine watersheds are also sensitive indicators of changes in atmospheric deposition because there is often very little vegetation or soil to dampen the impacts of changes in the magnitude or chemical composition of atmospheric deposition. Here we have demonstrated that alpine lakes may present valuable geochemical archives for reconstructing changes in atmospheric deposition. Lakes in the SJM of southern Colorado indicate that dust deposition has increased between 300 and 500%. Not only does this dust have an important direct impact on the radiative budget of the atmosphere (Winckler et al., 2008) and the snowpack (Painter et al., 2007), it is also an important vector for transporting nutrients. Dust, as a nutrient vector, has likely depleted disturbed arid ecosystems of essential nutrients while subsidizing adjacent alpine ecosystems with essential nutrients. Here we show that increased dust loading has likely lead to the enrichment of alpine watersheds in important nutrients, such as N and P, base cations, such as Ca and Mg, and also certain metals, such as Cr, Cu, and Ni. Although this increase in dust loading appears to be regionally coherent over the SJM, local properties, such as watershed dimensions and underlying bedrock geology may amplify or dampen the ecosystem response to these atmospherically derived nutrient subsidies.

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Table 1. Trace element concentrations and isotopic compositions of sediments, dust, and bedrock. There was no significant difference between sediment and bedrock samples for various watersheds in the San Juan Mountains, therefore these samples have been pooled for all analyses. All values are reported with their respective standard deviations in parentheses. These are the values used in our Bayesian mixing model (Eq. 1).

Substrate	$^{87}\text{Sr}/^{86}\text{Sr}$	Sr (ppm)	$^{143}\text{Nd}/^{144}\text{Nd}$	Nd (ppm)	$^{147}\text{Sm}/^{144}\text{Nd}$	Sm (ppm)
Sediments	0.71609 (0.00109)	104.06 (17.95)	0.51209 (0.00001)	19.77 (2.84)	0.10306 (0.00260)	3.32 (0.44)
Dust	0.71632 (0.00301)	114.98 (25.67)	0.51210 (0.00001)	13.86 (1.06)	0.10775 (0.00171)	2.47 (0.21)
Bedrock	0.70797 (0.00587)	252.34 (225.25)	0.51222 (0.00005)	40.87 (9.40)	0.11429 (0.01255)	4.05 (5.60)

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Table 2. Statistical relationships between organic carbon flux and nitrogen and phosphorus availability. All statistical models are general linear models of the form $C = mN + mP + b$, where the response in organic carbon (C) is expressed as a function of organic nitrogen (N) and phosphorus (P) availability and the slope coefficients (m) are reported for both nutrients. Significant relationships are reported as P -values $< 0.0001 = ***$ and P -values $< 0.001 = **$, with their respective R^2 values. For multi-variate regressions the multiple R^2 value is reported. Akaike's Information Criteria (AIC) is also reported to evaluate the goodness of fit when additional model parameters are added.

Lake Basin	Nitrogen	Phosphorus	AIC	R^2
Porphyry:	6.98***		55.36	0.914
		44.91***	118.89	0.701
Senator Beck:	6.32***	5.74	55.35	0.919
		15.32***	-60.19	0.956
	7.10***	3.32**	23.33	0.335
			-68.51	0.968

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Table 3. Watershed Mass Balance for Phosphorus loading to the two alpine watersheds investigated in this study. Values reported with standard deviations (\pm) when available.

Watershed Parameters	Alpine Watersheds	
	Porphyry	Senator Beck
Physical Parameters		
Lake Surface Area (m^2)	6920	110
Watershed Area (m^2)	165 910	1420
Dust Flux ($\text{g m}^{-2} \text{yr}^{-1}$)	~ 10	~ 10
Bedrock Weathering ($\text{g m}^{-2} \text{yr}^{-1}$)	~ 3	~ 3
Chemical Parameters		
P concentration in Dust ($\mu\text{g g}^{-1}$)	902 ± 138	902 ± 138
P concentration in Bedrock ($\mu\text{g g}^{-1}$)	288 ± 29	1111 ± 163
N concentration in Dust ($\mu\text{g g}^{-1}$)	4400 ± 600	4400 ± 600
N concentration in Bedrock ($\mu\text{g g}^{-1}$)	0	0
Flux Estimates		
P flux from Dust ($\text{mg m}^{-2} \text{yr}^{-1}$)	225	125
P flux from Bedrock ($\text{mg m}^{-2} \text{yr}^{-1}$)	46	47
Total P flux ($\text{mg m}^{-2} \text{yr}^{-1}$)	271	172
Total N flux ($\text{mg m}^{-2} \text{yr}^{-1}$)	1099	611
Total N:P flux (molar)	9.0	7.9

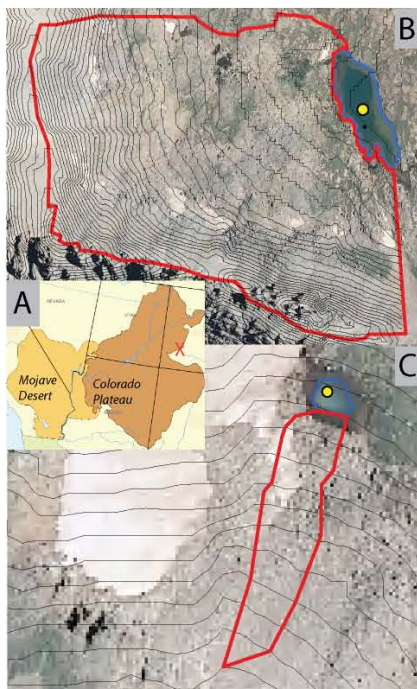


Fig. 1. Location of watersheds investigated in this study. The alpine watersheds investigated in this study are located in the San Juan Mountains (red X) directly downwind of the Mojave Desert and the Colorado Plateau (A). Lake sediment cores (yellow points) were collected from Porphyry (B) and Senator Beck (C) lake basins. The perimeter of each lake sampled is delineated in blue and the perimeter of each watershed is delineated in red. Contour lines (thin black lines) at 10 m intervals were used to estimate watershed area using ArcGIS (see methods). Green patches in orthophotos represent vegetation, grey patches represent talus slopes, and white patches represent seasonal snow cover with discoloration due to dust deposition. Note differences in scale of the two watersheds and morphometric parameters for each watershed reported in Table 3.

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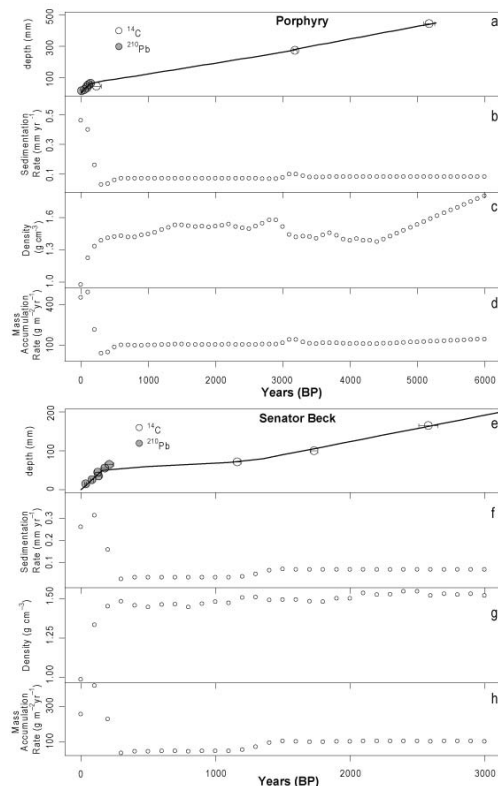


Fig. 2. Age-depth relationship of sediments from Porphyry (top panels) and Senator Beck (bottom panels) Lakes. Age estimates inferred from ^{14}C and ^{210}Pb for Porphyry (a) and Senator Beck (e). Sedimentation rates calculated as the first derivative of the spline functions fit to the age-depth relationships for Porphyry (b) and Senator Beck (f). Density measurements of sediments in Porphyry (c) and Senator Beck (g). Mass accumulation rates calculated as the product of sedimentation rates and density measurements in Porphyry (d) and Senator Beck (h).

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Total Sediment Fraction

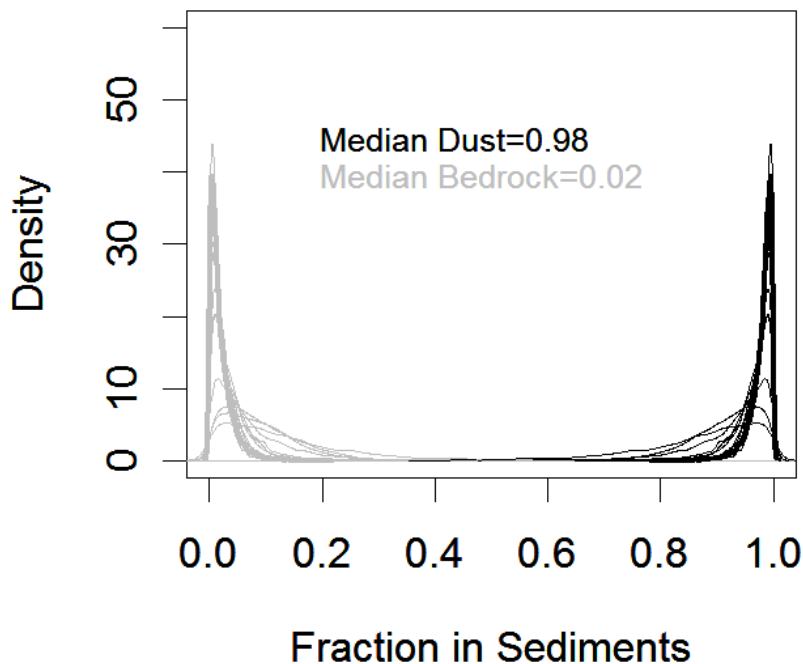


Fig. 3. Fraction of sediments derived from dust versus bedrock. Plotted are the probability density functions (PDFs) calculated from our mixing model. The thin lines correspond to the conventional two-end-member mixing models using all possible combinations of elemental concentrations and their isotopic values (Table 2). The thick lines represent the PDFs of sediment derived from bedrock versus dust when all isotopic tracers and their elemental concentrations are included in the mixing model. Based on this approach, approximately 98% of the sediment is derived from dust and only 2% from bedrock.

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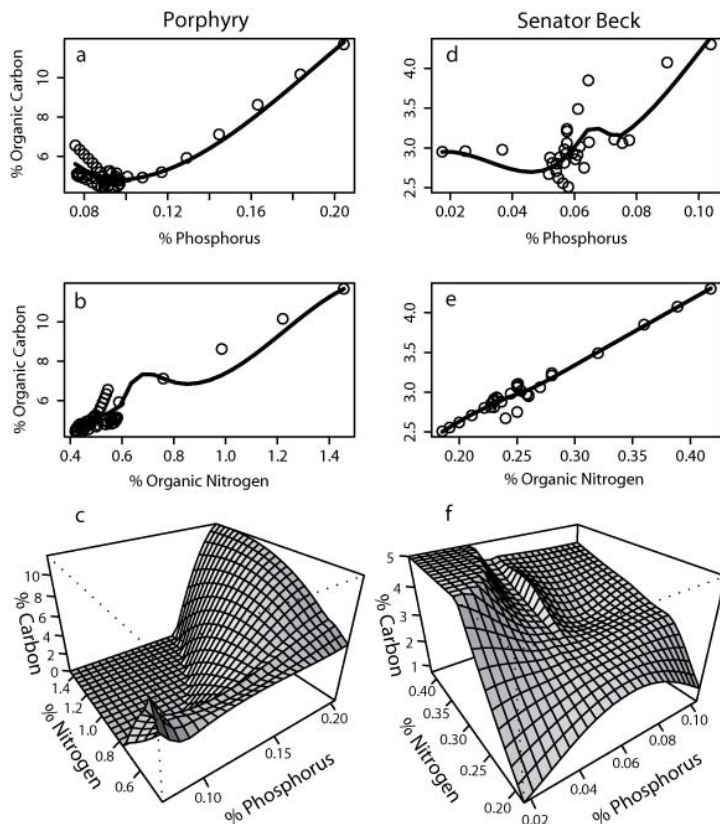


Fig. 4. The response of organic carbon content to nitrogen and phosphorus availability in the sediments of our two alpine lakes. The response of organic C to N and P in Porphry Lake (**a–c**) is compared to the response of organic C to N and P in Senator Beck Lake (**d–f**). Top panels (a, b, d, and e) show the response of organic C to N and P respectively and bottom panels (c and f) show the response surfaces of organic to N and P simultaneously. Note the higher C, N, and P content in Porphry as evidenced by the different scales.

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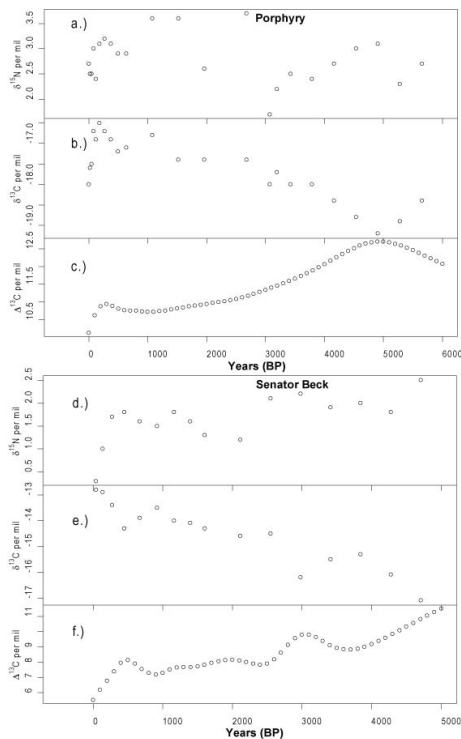


Fig. 5. Changes in the stable isotopic composition of carbon and nitrogen in lake sediments over time. Top panels (a–c) show changes in isotopic composition for Porphyry Lake sediments and bottom panels (d–f) show changes in isotopic composition of Senator Beck Lake sediments. Nitrogen isotopic ratios ($\delta^{15}\text{N}$) of the organic fraction is reported in panels (a) and (d). Carbon isotopic ratios ($\delta^{13}\text{C}$) are reported in panels (b) and (e). The isotopic discrimination ($\Delta^{13}\text{C}$) of organic C against atmospheric CO_2 (Francey et al., 1999) is reported in panels (c) and (f). All isotopic data are reported as per mil.

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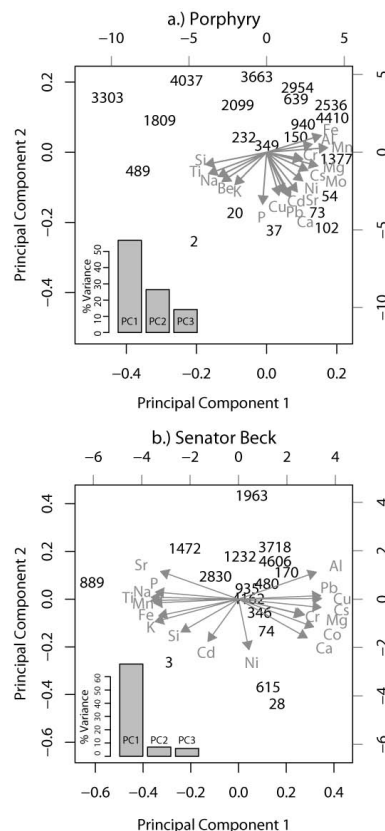


Fig. 6. Principal components analysis of elemental concentrations compared with dates plotted as years before present for lake sediments from Porphyry **(a)** and Senator Beck **(b)**. Bi-plots identify the variance in elemental concentrations (red arrows; top and right axes) that correspond with variance in age of sediments (black numbers; bottom and left axes). Inset grey bars in each figure show the amount of total variance in the array of data explained by the first 3 principal components (PC1, PC2, and PC3).

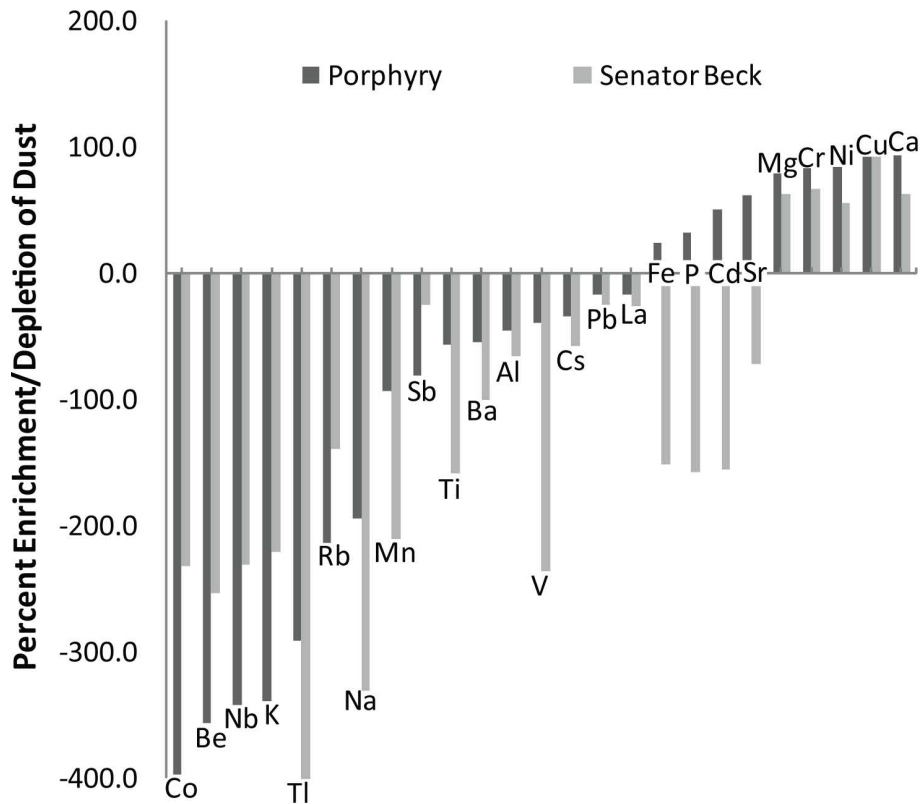


Fig. 7. Elemental concentrations of dust relative to local bedrock in the two watersheds. The elements that show greater concentration in bedrock relative to dust are shown as negatively depleted. The elements that show greater concentration in dust relative to bedrock are shown as positively enriched. Elements are plotted for both the Porphyry (dark grey) and Senator Beck (light grey) watersheds.

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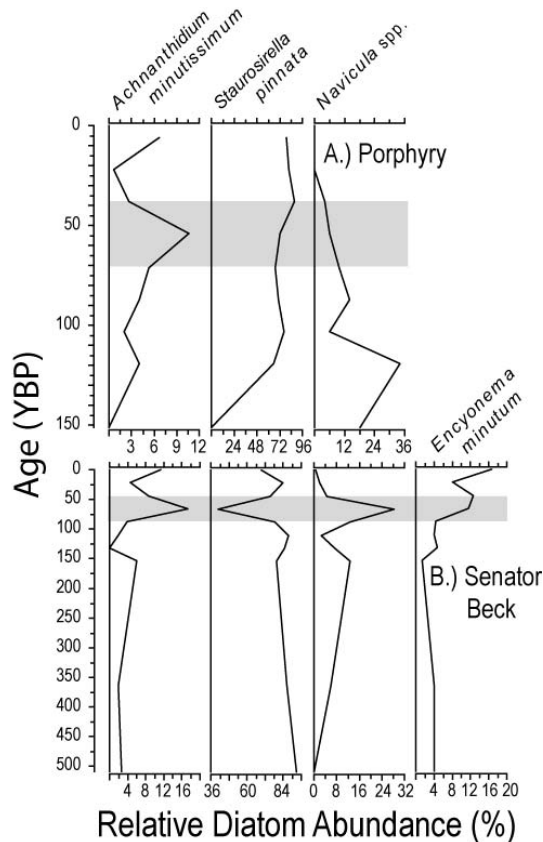


Fig. 8. Contemporary changes in the relative abundance of prominent diatom taxa in both Porphyry – **(A)**; top – and Senator Beck – **(B)** bottom – lake sediments. Diatoms are presented in relative abundance for the most prevalent diatom taxa, except *Navicula* spp. which represents the grouping of several different *Navicula* species. Intervals highlighted in grey represent periods of maximum dust accumulation.

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