Increasing eolian dust deposition in the western United States linked to human activity

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Mineral aerosols from dust are an important influence on climate and on marine and terrestrial biogeochemical cycles. These aerosols are generated from wind erosion of surface soils. The amount of dust emission can therefore be affected by human activities that alter surface sediments. However, changes in regional- and global-scale dust fluxes following the rapid expansion of human populations and settlements over the past two centuries are not well understood. Here we determine the accumulation rates and geochemical properties of alpine lake sediments from the western interior United States for the past 5,000 years. We find that dust load levels increased by 500% above the late Holocene average following the increased western settlement of the United States during the nineteenth century. We suggest that the increased dust deposition is caused by the expansion of livestock grazing in the early twentieth century. The larger dust flux, which persists into the early twenty-first century, results in a more than fivefold increase in inputs of K, Mg, Ca, N and P to the alpine ecosystems, with implications for surface-water alkalinity, aquatic productivity and terrestrial nutrient cycling.

Eolian dust is generated from a wide range of sources including industrial emissions and the wind erosion of soils¹. Dust may affect ocean productivity^{2,3}, control terrestrial nutrient cycling⁴ and alter regional and global climate^{5,6}. Dust deposition onto snow cover in the western United States has recently been shown to accelerate melt and reduce snow-cover duration by approximately one month, a finding that has broad implications for water resources in mountainous regions of the United States⁷. At a global scale, the Sahara and Sahel deserts in Africa and the deserts of central Asia produce most of the world's mineral aerosol load¹. Regional sources of dust, however, produce significant quantities of mineral aerosols with effects on soil fertility, air quality and human health^{8,9}. Given the wide range of potential impacts of atmospheric dust, it is critical to improve our understanding of the past and present role of human activities on dust emission and deposition.

The Great Basin, Colorado Plateau, Mojave and Sonoran deserts of the southwestern United States are responsible for the majority of emissions in North America¹. Like many arid environments, the drylands of the western United States have experienced widespread land-use change over the past two

centuries, with rapid acceleration of agricultural and grazing activities following the westward expansion of the United States in the 1800s (ref. 10). Despite growing evidence of the impacts of land use on wind erosion of soils around the world^{11–13}, the history of human influences on atmospheric dust remains poorly documented. Records showing increased dust accumulation in Antarctic ice cores between the nineteenth and twentieth centuries¹⁴, and evidence for changing chemistry of glacial dust during the twentieth century¹⁵, suggest higher contemporary atmospheric mineral aerosol loads than during the pre-industrial period. Similar conclusions have been reached in studies of peat bogs in Europe¹⁶. Without more documentation of contemporary and palaeo-deposition rates, however, we are largely limited to speculation about how humans have altered regional and global dust emissions.

DUST PROXY RECORDS

We obtained proxy records of dust deposition from high-elevation lakes in the San Juan Mountains in southwestern Colorado to

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Figure 1 Sediment accumulation rates and physical properties. a–**c**, Sediments from Porphyry Lake including sediment and mass accumulation rates (**a**), density (**b**) and texture (**c**). **d**–**f**, Sediments from Senator Beck Lake, including sediment and mass accumulation rates (**d**), density (**e**) and texture (**f**). The age scale is on the basis of the depth–age model described in the supplementary online material, with age dates in the top 150 yr determined from ²¹⁰Pb measurements and those below from ¹⁴C dating of terrestrial macrofossils. The error bars in **b** and **e** are estimated 95% confidence limits for density measurements.

examine the possibility of human-induced changes in atmospheric dust deposition. Located downwind of major western US deserts, this mountain range is prone to frequent eolian deposition events (see Supplementary Information, Fig. S1). The mountain range also contains a number of high-elevation lakes located above the tree line in areas with limited soil development. Given the frequent inputs of dust to snow cover⁷, these alpine lakes provide ideal locations for studying changes in dust loading.

In two alpine lakes, sediment accumulation rates over the past $\sim 150 \,\mathrm{yr}$ are more than five times greater than average accumulation rates over the past 5,000 yr, on the basis of radiogenic ²¹⁰Pb and ¹⁴C dates (Fig. 1). These recently elevated sedimentation rates are supported by an independent study on a larger, nearby subalpine lake that also shows large increases (7–17-fold) in recent- versus late-Holocene sedimentation rates¹⁷ (Table 1). Changes in sedimentation rates in these lakes occurred in the transition between age records derived from terrestrial macrofossils and those derived from ²¹⁰Pb (see Supplementary Information, Fig. S2). Although it is impossible to assign an exact date to the onset of increasing sedimentation rate, there is a clear and abrupt transition in sedimentation rate from old (\sim 1,000 yr before present) to recent (\sim 150 yr before present) sediment ages in one of our study lakes (Senator Beck Lake). The age and thickness of lake sediments also provide evidence that sedimentation rates peaked $\sim 100 \, \text{yr}$ before present. Taken together, these data strongly suggest that the period of increased sedimentation rate occurred within the past two centuries (Fig. 1, Supplementary Information, Tables S1,S2).

The San Juan Mountains currently experience four to seven large winter-time dust deposition events each year⁷, and the recent increases in lake-sediment accumulation could be caused by these periodic deposition events. To test this possibility, we examined the physical and isotopic properties of dust, bedrock and lake sediments. The lake sediments are dominantly composed of finegrained silts consistent with a far-travelled eolian origin rather than a local source¹⁸. To test further whether the sediment is dominantly dust derived, we compared the neodymium and strontium isotopic compositions of both the coarse- (>250 µm) and fine-grained $(37-63 \,\mu\text{m})$ lake sediments with those of the bedrock underlying their catchments. The range of $\varepsilon_{\rm Nd}$ values of ~ -5 to -9.8 from the bedrock (Oligocene silicic volcanic rocks of the San Juan volcanic field) underlying these basins contrasts with values from finegrained lake sediment that range from ~ -10 to -11, despite similar values of ¹⁴⁷Sm/¹⁴⁴Nd ratios from ~0.10 to 0.11 (Fig. 2a, Supplementary Information, Table S3). The ε_{Nd} values (from -10.4to -10.8) and ¹⁴⁷Sm/¹⁴⁴Nd ratios from dust, collected from San Juan snowpack in 2005, are indistinguishable from those in the fine-grained sediment in the lakes (Fig. 2a). Strontium isotopes show similar patterns, with ⁸⁷Sr/⁸⁶Sr ratios (<0.708) from the residual fraction of the local volcanic bedrock consistently lower than those from lake sediments (>0.713). As with the Nd isotopes, the $^{87}\text{Sr}/^{86}\text{Sr}$ ratios for the 37–63 μm fraction of sediments are nearly identical to values for contemporary dust (Fig. 2b). The combination of Sr and Nd isotopes and sediment texture provides unambiguous evidence that the fine fraction of lake sediments (>75% of typical sediment mass in these cores) is derived from exogenous sources rather than locally eroded bedrock.

Table 1 Estimates of sediment accumulation rates for lakes in the San Juan Mountains, Colorado. The Mineral Basin, Senator Beck and Porphyry Lakes were sampled for this study, whereas the Molas Lake data are from ref. 17. Sediment accumulation rates are presented for dated intervals where information is sufficient to estimate sedimentation rates. Values in parentheses are standard errors estimated from propagation of analytical uncertainties through age/depth regression equations.

Alpine lake	Dated interval	Dating method*	Sediment accumulation rate (mm yr ⁻¹)	Mass accumulation rate (g m ⁻²)
Mineral Basin Lake	\sim 1,890-present	²¹⁰ Pb	0.6 (0.06)	ND
Senator Beck Basin Lake	~1,910-present ~1,850-1,910 1,750-1,150 BP 2,580-1,750 BP	210Pb 210Pb ¹⁴ C ¹⁴ C	0.42 (0.04) 0.44 (0.04) 0.050 (0.001) 0.078 (0.001)	457 (18) 555 (20) 92 (10) 124 (2)
Porphyry Lake	~1,910 to present ~1,850 to 1,910 5,178–3,183 вР	²¹⁰ Pb ²¹⁰ Pb ¹⁴ C	0.45 (0.05) 0.47 (0.05) 0.092 (0.001)	501 (26) 576 (74) 119 (1)
Molas Lake ¹⁷	~1,880-present 1260–120 BP 2760–1260 BP 5920–2760 BP	²¹⁰ Pb ¹⁴ C ¹⁴ C ¹⁴ C	5.31 1.70 0.32 0.28	ND ND ND ND

* 202 Pb refers to the use of lead-210 dating techniques for recent sediments, whereas older sediments were dated through the measurement of the ¹⁴C content of terrestrial macrofossils.



Figure 2 Dust, bedrock and sediment isotopic characteristics. a, The ε_{Nd} versus ¹⁴⁷Sm/¹⁴⁴Nd data, which illustrate the separation between local bedrock, dust and sediment. The bedrock values are from the San Juan volcanic field (referenced data in main text) plotted for comparison with contemporary dust and two size fractions from lake sediments. **b**, The strontium isotope values for dust, bedrock and sediments in the study area. All values are for residual fractions of samples (see Supplementary Information). Analytical errors for both Sr and Nd isotopic measurements are smaller than the marker size in the graphs.

SOURCES OF DUST TO THE SAN JUAN MOUNTAINS

There are a number of potential sources of dust to the San Juan Mountains including the deserts of the southwestern United States and desert sources in Asia that are known to contribute dust to the North American continent¹⁹. Although the precise provenance of San Juan dust samples is difficult to determine, the physical and isotopic properties of dust can be used to substantially narrow the potential source regions. The $\varepsilon_{\rm Nd}$ values of contemporary dust range from -10.4 to -10.8 at $^{147}\rm{Sm}/^{144}\rm{Nd}$ ratios between 0.106 and 0.110. These values overlap those of Palaeoproterozoic basement rocks that comprise the bulk of the continental crust underlying Arizona and New Mexico (Fig. 3a, Supplementary Information, Table S3)^{20–27}. These isotopic compositions are

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Figure 3 Dust isotopic properties and texture. a, ε_{Nd} and 147 Sm/ 144 Nd for potential sources of dust to the San Juan mountains. Except where otherwise noted, data in the graph are for Precambrian basement rocks for various portions of the western United States as cited in the main text. Analytical errors are smaller than the graph symbols. **b**, The average grain-size distribution for modern dust collected from five separate deposition events in 2005. Bars on the graph represent the 95% confidence interval.

consistent with dust sources to the south and/or southwest of the study area. Satellite detection of dust plumes and atmospheric back-trajectory modelling for this region also link wintertime dust deposition in the San Juan Mountains to dust plumes that originate in the deserts of the southwestern United States, further supporting a dust source indigenous to western North America⁷.

Although Asian dust periodically falls on the San Juan Mountains, the textural distribution of dust samples also provides strong evidence for a regional source of dust. Nearly 40% of the mass of dust sampled from the snowpack occurs in the 10–37 μ m size class, 26% in the 37–63 μ m size class and 17% in the 63–180 μ m size class (Fig. 3b). The relatively large proportion of particles over 37 μ m is evidence for particles that have been transported hundreds, rather than thousands, of kilometres²⁸. This result suggests that the dominant source of wintertime dust inputs to the San Juan Mountains is the western United States rather than far-travelled Asian dust, which would be much finer (that is, in the less than 10 μ m size classes)²⁹.

CAUSES OF INCREASED DUST LOADING

Increased sedimentation in lakes in recent decades or centuries is generally attributed to catchment-scale disturbance³⁰ or changes in regional climate³¹. In the high-elevation setting sampled here, the lakes are surrounded by talus fields, with limited soil and vegetation development and no evidence of modern human disturbance. These factors together effectively rule out a local cause for the observed increase in lake sedimentation rates. Combined with the isotopic evidence for predominantly eolian sources for the lake sediments, we suggest that the large, recent increases in sedimentation rates are related to increased eolian deposition. Both lake-sediment core records indicate that increased dust loading began between 200 and 100 yr ago with peak deposition rates in the first half of the twentieth century (Fig. 1). Drought is a potential cause of increased wind erosion of soils. Although there have been multiple drought events during the nineteenth and twentieth centuries in the western United States, these droughts have been considerably shorter and less severe than several drought events that have occurred over the past 2,000 yr in the region³², making it unlikely that drought is the primary cause for the increased dust loading observed in this study. Instead, the period of increased sedimentation rate is contemporaneous with an intensification of western US land use, and particularly livestock grazing activities, that began in the early 1800s.

The migration of settlers of European descent into the western United States led to widespread expansion of grazing, mining and agricultural activities in the nineteenth and twentieth centuries. In the period following the development of railroad lines (and heavy transport capabilities for livestock) in the late 1860s, cattle and sheep grazing greatly intensified across the western United States^{10,33-35}. In the Navajo Nation tribal lands to the south and southwest of the San Juan Mountains, high animal densities and impacts of overgrazing became a major issue by the early 1890s. By the early 1930s, two-thirds of the land area in northeast Arizona had been significantly disturbed by heavy livestock use³⁶. Overall, nearly 70% of the natural ecosystems of the western United States have been affected by livestock grazing³⁷, resulting in loss of soil stability and increases in wind erosion of soil9,11. The extensive degradation of western US rangelands led to the Taylor Grazing Act of 1934, which imposed regulations and restrictions on grazing activities in



Figure 4 Elemental fluxes for two alpine lakes in the San Juan Mountains, Colorado. a–**c**, C, N and P fluxes, respectively, for Senator Beck and Porphyry Lakes. **d**–**f**, Ca, Mg and K loading, respectively, for Senator Beck and Porphyry Lakes. Element accumulation rates were calculated by multiplying mass accumulation rates by sediment elemental abundance and are expressed on an area basis. Error bars are on the basis of the propagation of error in density and element abundance measurements. Sediment ages for the top 150 yr were determined from ²¹⁰Pb measurements and those below from ¹⁴C dating of terrestrial macrofossils.

these rangelands. At about this time the mass accumulation rates of the lake sediments begin a moderate decline, which persists through the second half of the twentieth century (Fig. 1).

BIOGEOCHEMICAL IMPLICATIONS OF INCREASED DUST DEPOSITION

Eolian dust mobilized from arid-land soils generally contains high concentrations of base cations, and dust typically has high concentrations of N and P, as well as elevated concentrations of a range of atmospheric pollutants^{4,38}. High-elevation lakes and tundra ecosystems are generally low in nutrient content and vulnerable to increases in atmospheric deposition³⁹. There is strong evidence for the impacts of changing N deposition in highelevation settings⁴⁰, as well as suggestions of increasing P and basecation deposition into high-elevation settings^{39,41,42}.

To evaluate the changes in element loading in the lake sediments examined here, we combined sediment mass-accumulation rates with geochemical analyses to estimate sediment elementaccumulation rates in the two lakes. For the elements N, P, Ca, Mg and K, both lakes show large increases in element accumulation rates over the past 150 yr compared with background fluxes (Fig. 4). Element accumulation rates for the base cations, Ca, Mg and K, generally show a peak $\sim 100 \, \text{yr}$ ago, with some decline to present and some variation (particularly with Mg) between the lakes. These early-twentieth-century peaks provide additional evidence that land-use change in the western United States led to a large destabilization of base-cation-rich, desert soils in the early twentieth century. There is evidence from a range of other settings that base-cation loading via dust deposition can change precipitation and surface-water alkalinity43,44. The relatively large perturbation to base-cation loading to these lakes suggests that

dust inputs could be one factor mitigating the lake impacts of generalized regional increases in acid deposition⁴⁵.

Both lake-sediment records show an increase in C and N accumulation rates during the nineteenth and twentieth centuries. These records are complicated to interpret, both because of some variability between the lakes and because changes in the accumulation of these elements could be a product of either diagenesis in the sediment profile⁴⁶ or increases in aquatic productivity resulting from elevated dust and nitrogen deposition. For Porphyry Lake, the sediment N and organic C record shows increasing accumulation through the twentieth century, although this is less pronounced in Senator Beck Lake. Nitrogen deposition to alpine settings is the result of multiple potential sources, probably dominated by gas-phase emissions of nitrogen oxides during fossil-fuel combustion⁴⁷. The timing and magnitude of N deposition changes to these lake sediments is generally reflective of global perturbations to the N cycle⁴⁸ although both lakes also show peaks in C and N accumulation that are consistent with the apparent dust deposition peak \sim 100 yr ago and suggest a potential stimulatory effect of this dust on lake productivity.

Phosphorus sedimentation accumulation rates show a large and sustained increase through the modern record in both cores, which indicates a more complex change in element loading than observed for the base cations. Phosphorus, like the base cations, shows elevated element accumulation rates early in the twentieth century in addition to more recent increases in loading rates. Phosphorus deposition is also subject to the diagenesis issues for C and N mentioned above. Nonetheless, the continued increase of P sedimentation rates through the twentieth century is contemporaneous with the widespread expansion of the use of P fertilizers in the second half of the twentieth century⁴⁹. This increase is caused by both elevated rates of sediment accumulation and increasing P concentrations in sediments. The possibility of human disruption to the P cycle through agricultural activity and dust emission is a potentially significant finding, as relatively little is known about anthropogenic changes in P deposition rates to ecosystems. Although intriguing, more work is needed to establish the causes of these changes in P accumulation rates.

CONCLUSIONS

Land-use change in the western US over the past 200 yr is similar to agricultural intensification in semi-arid regions around the world, which often results in increased wind erosion of soils^{12,14,50}. Recent studies of dust deposition records from Antarctica¹⁴ suggest that the patterns observed in our study may be replicated elsewhere and may be indicative of a significant human role in regional dust generation. If human land-use change has altered the flux of dust between the biosphere and the atmosphere to the degree implied by our study, then there have also been substantial changes in elemental fluxes to ecosystems, with broad implications for nutrient deposition and biogeochemical cycling. In addition to its role in nutrient and contaminant transport, dust can also influence regional snowpack and climate. Effects on snowpack include accelerated spring snowmelt and decreased late-spring snowpack depth⁷. In the atmosphere, mineral aerosols play an important and highly uncertain role in climate change⁵¹. Most studies of mineral aerosols focus on the less than 10 µm size classes generated in, and transported from, Asia and Africa. The results of this study suggest the importance of regional sources for mineral aerosol fluxes in the western United States, and imply a strong and changing human role in controlling these fluxes. To the degree that these results are replicated in other areas with extensive, recent land-use change, human-caused changes in dust production and deposition may be far more important than previously thought.

METHODS

The alpine lakes sampled in this study are located between Silverton and Telluride, Colorado, USA. The lakes were located at \sim 3,500 m in alpine glacial cirques with shallow surrounding soils and limited vegetation development (grasses and small shrubs). Sediment cores were extracted using a Universal Core Head Corer from shallow alpine lakes (1–2 m depth). In the laboratory, the cores were subsampled into 0.5–1 cm increments. Terrestrial macrofossils were picked from the sediments during this initial sectioning procedure. We obtained as many macrofossils as possible for this study; however, these samples are limited owing to the high elevation and limited vegetation cover. All samples were freeze-dried and separated using a splitter to yield homogenous samples. Dust samples were collected from snowpack within 2–3 weeks of deposition in Senator Beck Basin. These samples were melted, evaporated and freeze-dried. Several samples of representative bedrock were also taken from Senator Beck Basin.

Surface sediments from lakes in all basins were dated using the radiogenic nuclide ²¹⁰Pb, and sediments at depth were dated by measuring ¹⁴C of macrofossils. The sediment accumulation rate was estimated using the constant-rate-of-supply model fitted to measurements of unsupported ²¹⁰Pb activity⁵² (see Supplementary Information, Table S1 and Methods). For each sediment core, we present two periods of accumulation related to the time intervals from the mid-1800s to early 1900s and from the early 1900s to the present. Our selection of these intervals enables the use of at least three ²¹⁰Pb measurements per period, with the intent of providing a relatively conservative estimate of recent temporal variation in sediment loading. The surface sample from Porphyry Lake was excluded from the sedimentation estimates because of a suggestion of disturbance of the top 1 cm during sampling. Inclusion of this sample point doubles recent sedimentation estimates, but with sufficient uncertainty to warrant a more conservative approach.

Terrestrial macrofossils, identified as roots or woody material, were sampled in Senator Beck and Porphyry lake sediments for ¹⁴C analysis. Each

macrofossil was sonicated and then freeze-dried. The samples were analysed for ¹⁴C at the UC Irvine Keck Accelerator Mass Spectroscope facility (see Supplementary Information, Table S2). An oxalic acid standard of known ¹⁴C age was also freeze-dried and analysed for ¹⁴C content, yielding values within the range of analytical uncertainty, indicating that this method produces accurate radiocarbon ages. A depth-to-age model was constructed using the ²¹⁰Pb- and ¹⁴C-based chronologies (see Supplementary Information, Fig. S2), and this model was used in combination with density and chemistry measurements described below to estimate mass and elemental accumulation rates through time. The offset between ²¹⁰Pb and ¹⁴C records make it impossible to precisely constrain the timing of increased sedimentation; however, the record from Senator Beck Lake suggests that this period probably began in the 200 yr before present (see Supplementary Information)

The density and texture of each core were also measured using methods described in the Supplementary Information. The elemental content of bedrock, sediments and dust were determined using inductively coupled plasma atomic emission spectroscopy and inductively coupled plasma mass spectroscopy. Samples were dissolved in HF using a microwave-assisted digestion method. Two bedrock standards and one soil standard were also analysed, yielding values within 5–7% of the expected values. We analysed five duplicate samples across the two cores, with the following coefficients of variation: Ca, 16%; K, 6%; Mg, 8%; P, 7%; Fe, 3%; Si, 13%. Relative elemental concentrations were measured for a duplicate core from Porphyry Lake using an EDAX Eagle III X-ray fluorescence analyser following methods described in the Supplementary Information. Carbon and nitrogen content was measured with a CNS combustion analyser. Carbon and nitrogen measurements were accurate to \sim 1%, with an average variance on duplicate samples of less than 1%.

Measurements of Sr, Sm and Nd were made on dust, bedrock and sediment samples from Senator Beck Basin. Samples were dissolved in concentrated HF and HClO4 following removal of ammonium-acetate-soluble materials. For sediments, we analysed two distinct size fractions, 37-60 µm and over 250 µm. Strontium was separated from the solution using SrSpec resins, whereas Sm and Nd are obtained using reverse-phase chromatographic techniques⁵³. Isotope dilution concentration and Sr and Nd isotopic determinations were obtained using a Finnigan-MAT 261 thermal ionization mass spectrometer. Isotope dilution concentrations were accurate to \sim 1% for Sr, and 0.5% for Sm and Nd. Total procedural blanks averaged \sim 1 ng for Sr, and 100 pg for Nd, during the study period. Thirty measurements of SRM-987 during the study period yielded mean 87 Sr/ 86 Sr = 0.71032 ± 2. Analyses were corrected to the SRM-987 value of 0.71028. Measured 143 Nd/ 144 Nd were normalized to 146 Nd/ 144 Nd = 0.7219. Thirty-three measurements of the La Jolla Nd standard during the study period yielded a mean 143 Nd/ 144 Nd = 0.511838 ± 8 (2- σ mean). ε_{Nd} $\varepsilon_{\rm Nd} = \{[(^{143}\rm Nd/^{144}\rm Nd)(\rm sample)/(^{143}\rm Nd/^{144}\rm Nd)(\rm CHUR)] - 1\} \times 10^4)$ values were calculated using a present-day 143 Nd/ 144 Nd(CHUR) = 0.512638 (see Supplementary Information, Table S3).

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Author contributions

All authors commented on the manuscript and participated in the analysis of project results. J.C.N. and A.P.B. designed and carried out the study and J.C.N. developed the paper. G.L.F. carried out the isotopic analysis and aided in the interpretation of sediment isotopic chemistry. N.M.M. provided assistance in evaluating project results in comparison with global patterns in dust deposition. J.L.C., J.T.O., C.R.L. and R.L.R. analysed sediment core and dust chemistry. T.H.P. and C.C.L. developed and implemented dust sampling protocols for snowpack and assisted in sampling.

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