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Key Points:
- Dust deposition in New Zealand increased in the twentieth century as compared to the last 2,000 years
- Dust deposition decreased from 1990 AD to the present
- Reconstructed dust fluxes appear to vary in response to historical land use changes in Australia

Supporting Information:
- Supporting Information S1

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Increased Dust Deposition in New Zealand Related to Twentieth Century Australian Land Use

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Abstract Mineral aerosols (dust) generated in the dryland regions of Australia have the potential to reach New Zealand through atmospheric transport. Although a large portion of dust in New Zealand originates in Australia, little is known about how dust deposition has varied over time in New Zealand or what may have caused this variation. We used geochemical dust proxies to examine the recent history of dust deposition to two alpine lakes in Kahurangi National Park, South Island, New Zealand. Geochemical indicators suggest that dust deposition began to increase around 1900, with the greatest deposition rates occurring from ~1920 to ~1990. In subsequent decades, dust deposition rates to New Zealand lakes appear to have declined. This rise and fall of dust deposition recorded in New Zealand lakes is consistent with dust records from the Antarctic Ice Sheet, Eastern Australia, and incidents of low visibility due to dust events recorded at Australian climate stations. The dust deposition rate over time also follows the temporal pattern of land use in south and central Australia over the time scale of the twentieth century suggesting a causal linkage. It is possible, and perhaps likely, that drought cycles also affected both emissions and transport pathways but over shorter time periods this was difficult to discern at the temporal resolution of these lake sediment cores. The increase in dust deposition to the high-elevation regions of New Zealand likely has implications for the biogeochemistry of alpine lakes in the Tasman Mountains.

1. Introduction

Anthropogenic activities have led to increases in the amount of material transported through the atmosphere, specifically gases, volatile heavy metal oxides, and particulates. This increased atmospheric flux has wide ranging implications for environmental processes and biogeochemical cycles (Mahowald et al., 2017). The relative isolation of Australia and New Zealand, combined with the well-defined arrival of European settlers in 1788, provides an advantageous setting for evaluating land use effects on long-range atmospheric transport and deposition. European settlement of Australia has fundamentally altered the landscape through pastoralism, agriculture, industry, and mining operations (McAlpine et al., 2009; Mudd, 2007; Pearson & Lennon, 2010). These changes have resulted in well documented increases in both anthropogenic emissions from combustion and dust emissions from erosion (Marx, McGowan, et al., 2014)

Approximately, 1/3 of Australian soils (900,000 km^2) are currently affected by wind erosion (Leys & Eldridge, 1998; Pickup, 1998). Dust emissions from the arid to semiarid landscapes of Australia are well documented and affected by natural changes over long- and short-term climate cycles (Hesse, 1994; Hesse & McTainsh, 2003; Petherick et al., 2009; Speer, 2013). However, soil erosion, resulting in dust emissions, is generally exacerbated by land degradation associated with land use activities, for example, intensive livestock pasturing, agriculture, roads, and urban uses (Baddock et al., 2011; McTainsh et al., 1990). More than half of the natural landscape of Australia has been cleared with southeast Australia being particularly affected in that less than 10% of native vegetation cover remains (McAlpine et al., 2009). This massive expansion in land use may have led to increased soil erosion and dust generation but information on the link between historical land use change and dust production at the continental scale is limited.

Land clearing began in the nineteenth century and increased after World War II primarily for livestock and cultivation and to a lesser extent mining (McAlpine et al., 2009). All three practices are known to destabilize soil surfaces and produce fugitive dusts with the appropriate climatic conditions (e.g., Silvester et al., 2009).

Livestock, especially hooved animals, and agricultural practices disrupt the natural biogenic and physical soil crusts that often form in arid regions (Belnap, 1995). These crusts serve to resist the erosive stress caused by wind, significantly reducing soil emissions and dust formation (Belnap & Gillette, 1998). Studies on Australian soils have found that the removal of soil crusts increased dust emissions by up to 700% (Leys & Eldridge, 1998). Further, in the absence of a soil crust, the threshold wind velocity needed to produce dust decreased from wind speeds that rarely occur (e.g.,63 km/h) to wind speeds that frequently occur (e.g., 21 to 30 km/h; Leys & Eldridge, 1998).

Dominant westerly winds transport eroded Australian soils, which are then deposited in western and southern Australia, the South Pacific, and western regions of New Zealand (McGowan & Clark, 2008; P. D. Neff & Bertler, 2015). Knight et al. (1995) estimated that some 3–7 million tons of dust are transported to the South Pacific every year. In one 1987 event, Knight et al. (1995) estimate that 1.7 to 3 million tons of dust traveled as far as New Zealand. However, contemporary changes in the deposition of Australian dusts in New Zealand associated with land use histories have not been previously evaluated.

In this study, we used alpine lake sediments as archives of exogenous material flux to western New Zealand through time. Alpine lake catchments in the northwestern region of the South Island of New Zealand are primarily underlain by granitic bedrock, whereas Australian dust producing regions (the Murray Darling and Lake Eyre Basin) are composed of lake sediments, alluvium, limestone, and volcanics (Alley, 1998; Kingham, 1998). Thus, exogenous dusts from Australia should be geochemically distinct from endogenous bedrock in New Zealand (Marx et al., 2005a; McGowan et al., 2005). In addition, alpine lakes are often excellent recorders of atmospheric deposition because their waters are naturally dilute and have larger airshed to watershed ratios than lower elevation lakes, and mountain ranges act as natural barriers to atmospheric transport and enhance both the wet and dry deposition of material through orographic precipitation (Catalán et al., 2006). Further, alpine catchments are typically steep with weak soil development, allowing for the efficient transfer of dusts from the catchment to the lake basin and ultimately the sediments (Brahney et al., 2014).

2. Materials and Methods

2.1. Site Description

Because dust deposited in New Zealand may be Australian or local in origin, we selected sites that would minimize the influence of local dust sources. Local sources include the many braided glaciofluvial systems that drain on the eastern (leeward) side of the Southern Alps (McGowan et al., 1996). To avoid sites that might capture local dusts, we selected lake basins in Kahurangi National Park on the windward side of the Tasman Mountains located on the northwest tip of the South Island (Figure 1). This region is upwind of the prominent New Zealand glaciofluvial dust sources and is geographically disposed to receive dusts and other aerosols from Australia (McGowan & Clark, 2008; Putman, 2017)

Lakes in the region of the Kahurangi National Park are ideal for examining far-travelled dusts due to two additional important characteristics. First, the lakes are situated in granitic basins, which is necessary for the separation of the Australian dust that originate from sedimentary bedrock from the local bedrock as these rock types are typically geochemically distinct. Moreover, Australian dust sources are geochemically different from the glaciofluvial sediments of the Southern Alps (Marx et al., 2005a; Marx et al., 2005b; McGowan et al., 2005), making geochemistry a useful tool in distinguishing local from far-travelled dust. Second, high alpine lakes within this watershed are remote and not likely to have been impacted by large-scale human perturbations or grazing (McIntyre, 2007; NZ Department of Conservation, 2010).

Two alpine lakes within the Tasman Mountains of the Kahurangi National Park were chosen for analysis; these are Lake Clara and Adelaide Tarn (Figure 1). Lake Clara is a small, 17.8-m deep headwater lake situated on a small rocky shelf at 1,332-m above sea level (masl) and has a catchment area of 0.35 km^2 and a lake area of 0.05 km^2 (CA:LA = 7). There are no inlet streams to Lake Clara. Adelaide Tarn is in an adjacent drainage at 1,256 masl, with a much larger catchment area of 1.8 km^2 and similar lake area of 0.06 km^2 (CA:LA = 31) a maximum depth of 7.1 m, and small inlet stream. Excluding precipitation, there is a lack of long-term climate measurements near the catchments studied. Predictions made by Leathwick (2002) estimate mean annual temperature as 5.7 °C for Lake Clara and 6.1 °C for Adelaide Tarn. Annual minimum

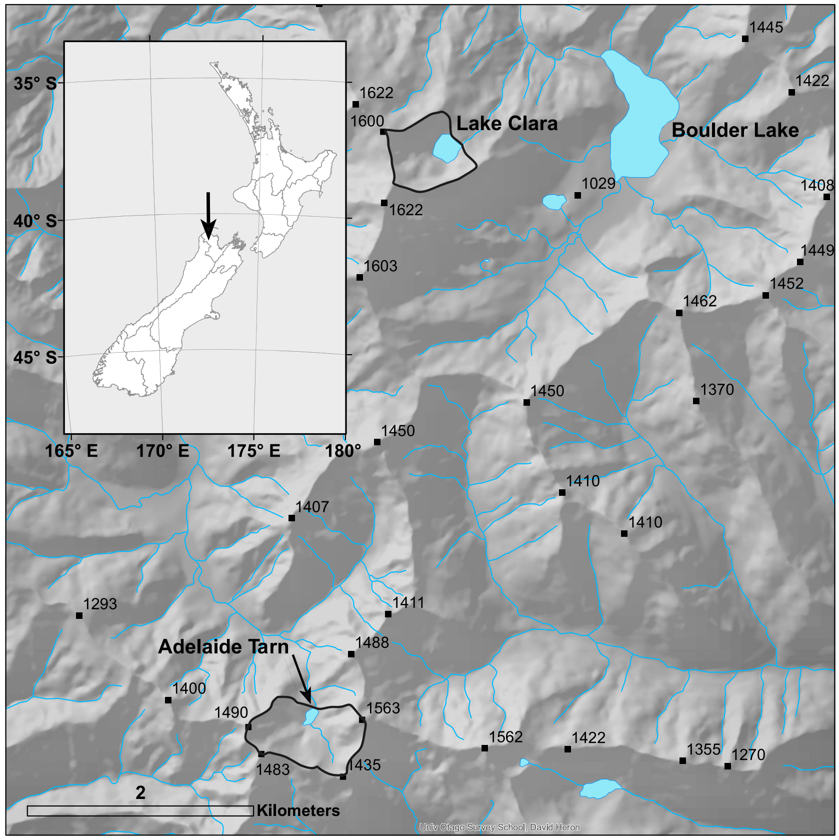

Figure 1. Kahurangi National Park, South Island, New Zealand. Drainage areas for both Lake Clara and Adelaide Tarn are delineated in black.

temperatures of about −2 °C suggest that periods of snow and ice accumulation occur but are not extensive. Average summer precipitation appears to be 30% lower than winter precipitation.

2.2. Sediment Core Analyses

Sediment cores were extracted from the deepest area of the pelagic region of each lake using a gravity coring system. A 22-cm core was retrieved from Lake Clara and a 39-cm core from Adelaide Tarn. Cores were transported to the GNS Science in Lower Hutt where they were described, sectioned, and analyzed for water content and density. Sediment cores were dated using both ^{210}Pb and radiocarbon analyses. Lead-210 analyses were conducted at MyCore Laboratories in Ontario Canada; sediment interval ages were calculated assuming a constant rate of supply of unsupported ^{210}Pb (Appleby & Oldfield, 1978) after determining that the Constant Initial Concentration model was inappropriate due to variable within lake sedimentation rates (Appleby, 2001). Radiocarbon analyses were conducted by accelerator mass spectrometry at GNS Science, Rafter Radiocarbon Laboratory, Lower Hutt, New Zealand. Fossil plant materials for radiocarbon analyses were retrieved from four depths in Adelaide Tarn and three depths in Lake Clara (Table 1). Age-depth models were constructed from both ^{210}Pb and Radiocarbon ages using BACON, a Bayesian age-depth modeling software (Blaauw & Christen, 2011). For Lake Clara, seven ^{210}Pb and three radiocarbon ages were used in the model, for Lake Adelaide six ^{210}Pb and four radiocarbon ages were used in the model. SHCal13 was used for radiocarbon calibration within the BACON model (Hogg et al., 2013).

To isolate the immobile mineral trace element fraction in the sediment cores, we used a two-tiered geochemical leaching method to remove trace elements associated with the organic and oxyhydroxide fractions that play a role in the sediment mobility of reactive metals. Step one consisted of a tetra-sodium pyrophosphate ($Na_4P_2O_7$) extraction to remove organic-metal complexes, and step two used sodium-citrate/dithionite

Table 1
Conventional Radiocarbon Ages for Lake Clara and Adelaide Tarn Sediment Cores

Sample ID	Lake	Depth(cm)	Material dated	δ13C	Radiocarbon age (years BP)	Range ± (years)
NZA 40094	Adelaide	15	Plant material	−27.9	543	20
NZA 40095	Adelaide	20	Plant material	−24.8	446	20
NZA 40096	Adelaide	25	Plant material	−30.5	850	20
NZA 40097	Adelaide	30	Plant material	−31.9	1078	20
NZA 50576	Clara	11	Plant material	−27.7	600	18
NZA 40257	Clara	16	Plant material	−30.1	1,359	45
NZA 40093	Clara	20	Plant material	−26.9	1,772	20

[$(Na_3C_6H_5O_7)$ $(Na_2S_2O_4)$] as a reducing agent to remove amorphous Fe and Mn oxides (Ross & Wang, 1993). Between treatments, the residual sediments were washed with distilled water and aspirated three times before the subsequent digestion and final analysis. The remaining residual or immobile fraction represents an operationally defined mineral composition that is resistant to diagenetic change and selective weathering once deposited. This fraction can be effectively used to distinguish between sediment sources (Brahney et al., 2008). Trace element analyses on the residual fraction were conducted using an inductively coupled plasma atomic emission spectrometer and an inductively couple plasma mass spectrometer at the Ontario Geological Survey Geosciences Laboratory, Canada, which is an ISO/IEC 17025 accredited laboratory. Samples were digested using a standard multiacid digest and checked against certified standards (LKSD-3) and blanks. Blank values for elements used in the study were all below detect and the percent errors determined from standards were between 0.7% and 2%. Replicates were performed every five samples and the root-mean-square coefficient of variation (Stanley & Lawie, 2007) is provided at an element-by-element basis in the supplementary material. The range of root-mean-square coefficient of variation for the elements used in this analysis are 0.6% to 6.9% in Adelaide Tarn and 0.4 to 3.1% in Lake Clara, which are 1 to 3 orders of magnitude below the variation observed in the core. All details on QA/QC can be found in the supporting information.

2.3. Dust Deposition History

Several studies using dusts obtained from glaciers on the western slope of the Southern Alps have shown that Australian dusts can be separated from the local New Zealand bedrock based on trace element compositions and Rare Earth Elements (REEs) (Marx et al., 2005a, 2005b; Marx, Lavin, et al., 2014; McGowan et al., 2005). Similarly, Marx, McGowan, et al. (2014) determined which elements behaved conservatively from entrainment to deposition and post deposition. These included a suite of REEs, alkali, alkaline earth elements, and other metals that they used in a mixing model to derive dust deposition rates to a peat mire in southeastern Australia. Here we build on these earlier studies to evaluate dust deposition to our lake sediments using two complementary methods. Method 1 uses the historical variation in REEs to evaluate historical changes in the primary mineral composition of the lake sediments. Method 2 capitalizes on the end-member mixing approach developed by Christophersen and Hooper (Christophersen et al., 1990; Christophersen & Hooper, 1992; Hooper, 2003) that uses principal components analyses to assign end-member contributions to a given mixture, in this case, a sediment interval. Because the latter method can estimate a proportional contribution to a sediment interval, the data can be combined with sediment fluxes to estimate historical deposition rates through time. Taken together, these two methods provide two different approaches to the determination of the dust proportion in sediment cores.

2.3.1. Method 1 Average *z* Score of REEs

Sediment interval REE concentrations including Y, La, Ce, Pr, Nd, Sm, Eu, Gd, Tb, Dy, Ho Er, Tm, Yb, and Lu were mean-normalized and subsequently averaged to provide a historical overview of compositional changes throughout the sediment core. We excluded heavy metals primarily derived from anthropogenic activities including As, Cd, Cu, Hg, Ni, Pb, Sb, Se, Sn, and V. We choose REES to provide a historical perspective on land use and industrial activities, respectively, upwind on the Australian continent. Individual and composite profiles from both Lake Clara and Adelaide Tarn are determined based on the historical variation of REEs. Error for the composite profiles was calculated by the pooled standard error.

2.3.2. Method 2 End-Member Mixing Analysis (EMMA)

Method 2 uses EMMA and a constrained least squares approach to derive dust proportions within each sediment interval. This method is adapted from stream chemistry source separation techniques developed by Christophersen et al., 1990, Christophersen & Hooper, 1992, 2003) as well as sediment source separation techniques (Brahney et al., 2008; Bryan et al., 1969; Marx, McGowan, et al., 2014). The method uses principal components analyses to assign end-member contributions. Dust end-member REE composition is determined from four samples collected off glaciers in the Southern Alps of New Zealand that represent long-travelled dust from Australian sources (Marx et al., 2005a, 2005b; McGowan et al., 2005). Bedrock end-members are derived from two and three representative rocks samples collected from the alpine catchments of Lake Clara and Adelaide Tarn, respectively. We recognize that bedrock does not likely represent a true end-member composition as rock weathering can alter REE concentrations under certain weathering conditions. However, because these are headwater lakes, we were unable to obtain inlet stream sediment concentrations as has been done elsewhere (e.g., Brahney et al., 2008), leaving the bedrock composition as the best available approximation for local source material. Given that the catchments are small and of a uniform geology, we would not anticipate river derived sediment to have a different mineral REE composition than bedrock eroded and transported through other means. Moreover, our use of largely immobile elements as a tool for the determination of provenance should minimize any potential effects that may occur due to diagenetic reactions and/or differential rates of weathering across particular mineral fractions in the bedrock.

Principal component analyses were performed on mean-normalized sediment core REE data. We used several methods to evaluate the number of PC components, or potential dust and bedrock end-members, to retain. First, by definition, the sediment interval samples must be fully bounded by the dust and bedrock end-members in question when plotted in PC mixing space. Second, we use the Kaiser criterion, where components with eigenvalues greater than 1 are retained and finally the scree plot method where components are retained up to the "elbow" of a scree plot. Once the number of dimensions or potential sources are determined, we orthogonally project the potential sources into the sediment PC mixing space using the eigenvector coefficients. If the sediment intervals are bound by all potential end-members, the percent contribution of each end-member to the sediments can be calculated using a constrained least squares equation (Brahney et al., 2008). Because each end-member is based on several observations, we compute a mean and standard deviation of each end-member. From this, we calculated 1,000 possible combinations of source end-member contributions to each sediment interval and report the mean and standard deviation of the resultant dust contributions. Finally, we determined dust mass flux by multiplying derived dust concentrations (g of dust/g of sediment) by the sedimentation accumulation rate (g of sediment cm^{-2} yr^{-1}) at each interval. The dust flux mass as presented here is the catchment integrated and density normalized flux of dust to sediments and is not equivalent to an estimate of actual dust deposition rates to the land surface. As with Method 1, we further derive a composite dust flux record by averaging dust deposition histories from both Lake Clara and Adelaide Tarn with the error determined by a pooled standard error.

2.4. Australian Dust Emission Proxies

The historical patterns of land use change in Australia for pastoralism, agriculture, and mining all followed similar trends. Sheep rearing in Australia began in 1788 with a small flock of just 70 sheep (Pearson & Lennon, 2010). Commercial pastoralism grew rapidly and by the early 1800s there were approximately 100,000 sheep and by midcentury as many as 13 million. Sheep populations rose through the latter half of the nineteenth century until a drought in the 1860s caused a significant loss of sheep throughout the continent. The post drought industry again continued to grow exponentially, and at several points in the last 50 years, there have been upward of 170 million sheep in south and central Australia (ABS, A. B. of S, 2015). Agriculture followed a similar pattern where droughts in the first half of the twentieth century suppressed development, however, following the Second World War technological advances and higher demand allowed for the rapid expansion of Australian farms (ABS, A. B. of S, 1988). Mining in Australia has increased unabated since the late nineteenth century, with significant increases also occurring post the Second World War (Mudd, 2007). Over-grazing, agriculture, and settlement in other regions, such as the arid southwestern US, have resulted in increased dust deposition in the last two centuries above the late-Holocene average (Neff et al., 2008; Routson et al., 2016).

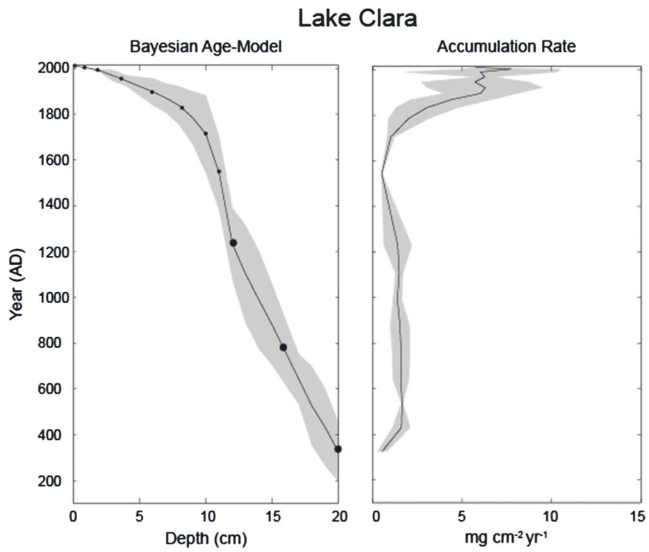

Figure 2. Lake Clara age model and sedimentation rates based on the Bayesian age model derived using BACON (Blaauw & Christen, 2011) and reported as AD. The 95% confidence intervals are shown as shaded grey areas. Radiocarbon sample points are represented by large black circles and ^{210}Pb points by small black circles.

Therefore, it is reasonable to hypothesize that an increase in extent and intensity of livestock grazing, agriculture, and mining in Australia has resulted in increased dust emissions.

Because data and statistics on annual livestock densities by Australian province are readily available, as compared to data pertaining to other potential dust sources such as mining and agriculture, we use total livestock numbers as an indicator for the timing of human agricultural activity that may have led to increased dust generation from Australia. Pastoralism is the dominant land use activity measured by areal extent and occurs along the same temporal trajectory as other human land use activities in Australia. Data were obtained from the Australian Bureau of Statistics's Historical Selected Agricultural Commodities (ABS, A. B. of S, 2015). Data include sheep, horse, and cattle population statistics totaled from New South Wales and South Australia to encompass the southern portion of the Lake Eyre Basin (where pastoralism dominantly occurs) and the Murray-Darling Basin. Sheep make up approximately 97 ± 1% of the total livestock numbers.

3. Results

3.1. Dating

The Bayesian age model for Lake Clara is based on three radiocarbon (Table 1) and six ^{210}Pb dates. No age reversals were found in either the ^{210}Pb or radiocarbon data for Lake Clara; however, a potential hiatus is suggested between 10- and 11-cm depth, around the midsixteenth century, where sedimentation rates drop significantly (supporting information Figure S1 and Figure 2). The sediments of this interval contained plant material suggesting lower lake levels during this period. During this interval, there is an apparent offset of ~350 years. In the sediment intervals prior, the age-depth relationships were approximately 19 years per centimeter of depth in the core. Excluding this interval, sedimentation rates in Lake Clara were relatively stable around of 1.5 mg cm^{-2} yr^{-1} from the start of the record (325 AD) to the midnineteenth century. Sedimentation rates rapidly increased through the middle to lake twentieth century peaking at 7.7 mg cm^{-2} yr^{-1} (Figure 2).

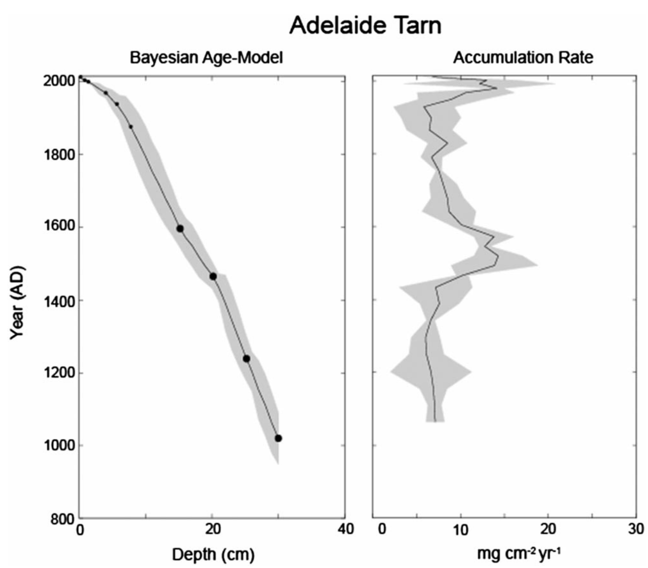

Figure 3. Adelaide Tarn age-model and sedimentation rates based on the Bayesian age model derived using BACON (Blaauw & Christen, 2011) and reported as AD. The 95% confidence intervals are shown as shaded grey areas. Radiocarbon sample points are represented by large black circles and ^{210}Pb points by small black circles.

The Adelaide Tarn Bayesian age model is based on four radiocarbon dates and five ^{210}Pb dates (Figure 3). No age reversals were found in the ^{210}Pb CRS age model; however, one age reversal was present in the radiocarbon ages and as a result, the sample was dropped from the model (supporting information Figure S2). The Adelaide Tarn datable record extends back in time to approximately 1000 AD. Sedimentation rates in Adelaide Tarn indicate two periods of elevated sedimentation from an average background rate of 6.6 mg cm^{-2} yr^{-1}. The first peak occurs from ~1470 to 1670 AD where sedimentation rates increase to a mean of 11.1 mg cm^{-2} yr^{-1}, and the second peak occurs from 1950 to 2002 AD, at a mean rate of 12.7 mg cm^{-2} yr^{-1} (Figure 3).

3.2. Sediment and Dust Geochemistry

The concentrations of REEs within dust samples is considerably greater than those measured within the catchment bedrock for both Adelaide Tarn and Lake Clara (supporting information) indicating that it is possible to use REEs to determine sediment provenance. REE profiles for both Lake Clara and Adelaide Tarn can be found in the supporting information Figures S3 and S4.

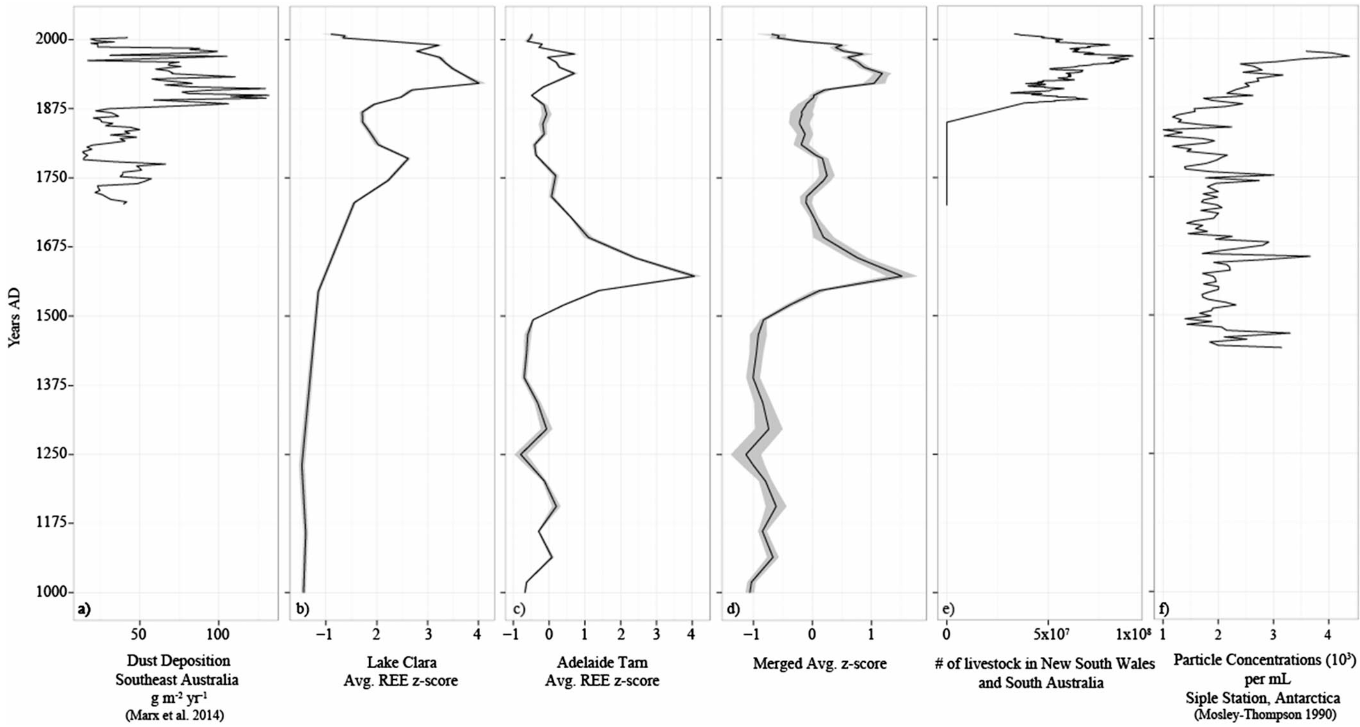

Figure 4. Historical profiles of (a) dust deposition as recorded by Marx, McGowan, et al. (2014) in eastern Australia, mean z score of REEs for (b) Lake Clara and (c) Adelaide Tarn, (d) the composite profile, (e) livestock densities for South and South-Eastern Australia presented here as a proxy measurement for the timeline of human land use intensification, and (f) dust deposition as recorded at the Siple Station in Antarctica (Mosley-Thompson, 1990).

3.2.1. Method 1 Average z-Score of REEs

Mean z-scored REEs for each lake sediment record and their composite record revealed a shift toward increased REE concentrations beginning in the late nineteenth to the early twentieth century (Figure 4). Adelaide Tarn also revealed an anomalous peak in some REEs at 16- to 7-cm depth (~1517–1615 AD). The peak is abrupt and up to 3 times background concentration. It is possible this peak is associated with a nearby volcanic eruption. Tephras dated in the Newall Formation near Mount Taranaki on the south end of the South Island indicated an eruption occurred at some time between 1500 and 1550 (Neall, 1972). The Egmont Volcano, or Mount Taranaki, has lava compositions that are enriched in the light REE (Price et al., 1992). Using elemental REE ratios, the Adelaide Tarn sediment anomaly plots along the mixing line toward the Egmont/Mount Taranaki Lava and away from dust and bedrock composition (Figure 5). This date corresponds temporally to the observed peak in the sediment record of Adelaide Tarn as well as the elevated particulate concentrations in the Siple Station ice core in Antarctica (Figure 4). Lake Clara did not indicate a similar increase in REEs, potentially due to the hiatus in the sediment record at this time. Peaks in REE concentrations in both cores occur around 1940 (±20), 1974 (±11.5), and 1990 (±5.5), numbers in brackets refer to the modeled standard deviation from BACON of as shown by shaded areas in supplementary Figures 1 and 2. Following the 1990 peak, values decline toward background concentrations near the surface of each core.

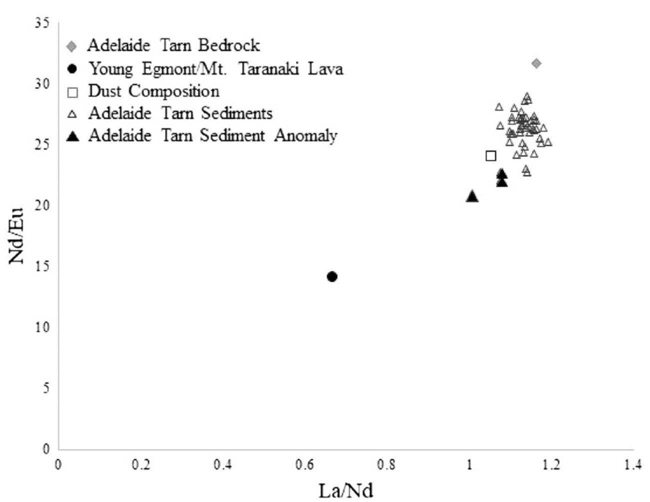

Figure 5. Rare earth element ratios for sediment, dust, Adelaide Tarn bedrock, and Mount Taranaki/Egmont young lavas. The sediment anomaly represented by filled triangles plots closer to lava end-member, suggesting this peak is likely due to ash deposition from an eruption of Mount Taranaki that occurred during this time.

3.2.2. Method 2 EMMA

Christophersen and Hooper (1992) determined that the number of end-members, or potential sources, that sufficiently explain the mixed

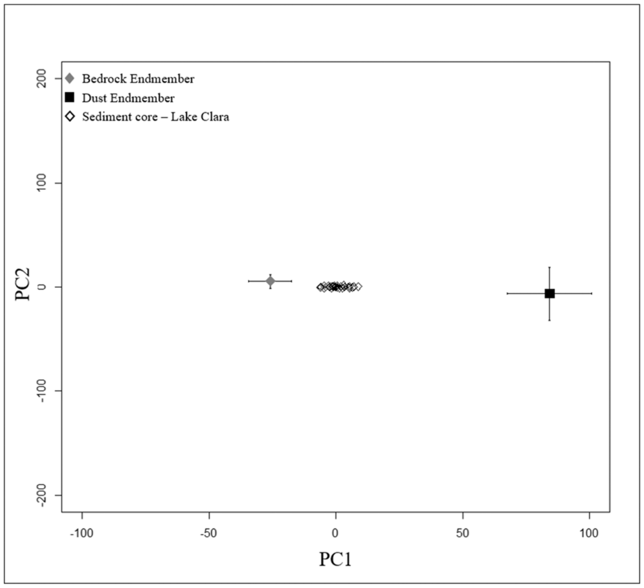

Figure 6. Principal component mixing space for Lake Clara showing sediments (open diamonds) and potential end-members, including dusts (filled square) and local bedrock (filled diamond), with lines representing the standard error. PC = principal component.

sample is equal to the number of Principal Components (PC) retained plus one. For the Lake Clara sediments, the Kaiser method (supporting information Table S1) and the scree plot method (supporting information Figure S5) indicate one principal component should be retained, explaining 87% of the variation in the data. Examining the data in mixing space (Figure 6) indicates that the sediment geochemistry is potentially explained by two sources, and the data is comfortably bounded by the local bedrock and dust geochemistry end-members. Therefore, we retain two end-members in the subsequent source partitioning equations. The Kaiser method for Adelaide Tarn suggests retaining the first three principal components, which explained 71%, 17%, and 7% of the variation (supporting information Table S1). This suggests that there are up to four end-member sources that could explain the data; at least one of these end-members is likely related to the anomalous peak in some REEs from 16- to 17-cm depth. The scree plot method, however, suggests retaining only one component indicating that 71% of the variation is explained in the first PC (supporting information Figures S6). Because we cannot acquire end-member geochemistry for the anomalous peak and because most of the variation can be explained using two end-members which together explain 88% of the variability, we retain only two potential sources in our partitioning equation. The sediments of Adelaide Tarn are fully bounded by the bedrock and dust geochemistry end-members (Figure 7).

The EMMA approach revealed similar historical variation as the z-score method (supporting information Figure S7). This result is not altogether surprising given that the same suite of elements was used. This approach, however, permits combining the derived end-member contributions with sedimentation rates to determine shifts in the historical flux of material to the lake sediments. In Lake Clara, proportional dust concentrations ranged from 0.19 to 0.34 and in Adelaide Tarn from 0.09 to 0.20. Differences in sediment dust concentrations between Lake Clara and Adelaide Tarn likely reflect differences in catchment properties, including catchment steepness, vegetation, and the catchment area relative to the lake area (CA:LA). These differences can influence the degree to which alpine lakes record atmospheric deposition by affecting the amount of dust retained in the terrestrial environment and transferred to the lake sediments versus dilution from catchment derived material (Ballantyne et al., 2010; Brahney, Ballantyne, et al., 2015; Morales-Baquero et al., 1999). In comparison to Adelaide Tarn, Lake Clara has a relatively small CA:LA (Figure 1). These properties, in general, would allow for greater dust transfer to the lake depositional area with less dilution from catchment-derived material. As noted above, it is important to distinguish the sediment deposition rates described here from true atmospheric dust deposition rates. Sediment deposition rates reflect a “catchment-integrated flux” because the flux to the sediment surface will include both the aerial flux to the lake surface as well as that mobilized from the catchment. Brahney et al. (2014) found that up to 40% of the dust material can be transported from the catchment to the lake basin, inflating the true atmospheric deposition rate. The degree to which dusts are mobilized from the catchment to the lake surface could, in principle, vary in time, and between lakes but such variation would have to be extreme to explain the results observed in this study and others using similar methodologies (Brahney, Ballantyne, et al., 2015; Neff et al., 2008; Routson et al., 2016).

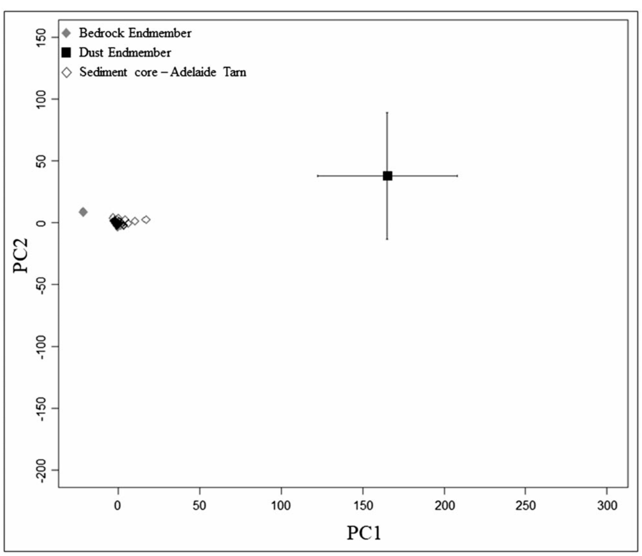

Figure 7. Principal component mixing space for Adelaide Tarn showing sediments (open diamonds) and potential end-members, including dusts (filled square) and local bedrock (filled diamond), with lines representing the standard error. Note that the error bars for the bedrock endmember are smaller than the symbol representing the mean. PC = principal component.

As with the z-scored data, EMMA analyses for both cores showed maximum inferred dust concentrations and deposition between 1920 and 2000 (supporting information Figure S7). The composite deposition record is shown in Figure 6 alongside other regional historical dust deposition data. The historical concentration of livestock in these areas shows a striking resemblance to the composite dust record from the Tasman Mountain region (Figure 8). Comparing the composite dust deposition estimate against livestock numbers, we find the two time series are highly correlated (r^2 of 0.78, $p < 0.0001$).

4. Discussion

Sediment geochemical records from two New Zealand lakes in Kahurangi National Park suggest that the loading of exogenous dust to this region of New Zealand increased beginning in the early twentieth century. Based on REEs concentrations in the local bedrock vs the Australian dusts collected from New Zealandglaciers, the likely source of dust within the lake sediment material is Australian in origin. Further, Neff and Bertler (2015) suggested that, if 5-day Hybrid Single Particle Lagrangian Integrated Trajectory model trajectories are representative of dust transport and deposition in New Zealand, then Australian sources may dominate over local dust sources, and that significant quantities of dust from other Southern Hemisphere regions are unlikely to reach New Zealand.

Shifts in sediment REE concentrations may result from differential mineral weathering or changes in the chemistry of exogenous dust. In these high elevation catchments, a shift in differential weathering and erosion leading to the observed patterns in cores would require an alteration of existing weathering and delivery processes in a way that was consistent through the twentieth century and which resulted in a dramatic change in REE concentration. One plausible explanation for such a shift is a climate-induced increase in the weathering and erosion of a specific bedrock fraction enriched in REEs in both alpine catchments. Such a shift could perhaps be explained by enhanced weathering due to elevated CO_2 (Clow & Mast, 2010); however, REE concentrations decline in the upper (recent) cores suggesting that increased differential weathering is an unlikely explanation for the patterns observed here. Instead, we suggest that the shift in REE concentrations reflects an increase in dust deposition derived from Australia. This is the most parsimonious explanation of sediment chemistry for two reasons. First, the measured REE concentrations from dusts (Marx et al., 2005a, 2005b; McGowan et al., 2005) are considerably greater than the mean bedrock composition. Second, these records align with several other proxies of dust mobilization and deposition in the region, including, the eastern Australian dust record as determined by Marx, McGowan, et al., 2014; Figure 4), dust records derived from Antarctic ice cores, and atmospheric models of Australian dust, all of which indicate increases in dust loading between 1930 and 1990 (Mahowald et al., 2010; Mosley-Thompson,1990). Dust concentrations from the Siple Station, Antarctica, that extend as far back as the fifteenth century reveal increased twentieth century dust concentrations, with post-1930 concentrations averaging $3.5 \pm 0.7 \times 10^3$ particles per ml (Figure 4). Peak concentrations occurred from 1950 to 1980 and background concentrations in the ice core averaged 1.7×10^3 particles per millimeter. Temporal variations in the ice core are similar to our reconstructed deposition rates (Figure 7).

The modern increase in dust deposition as determined from lake sediments is in accord with the elevated dust emission histories from the Lake Eyre Basin and other regions in Australia (DustWatch, 2015; McTainsh et al., 2007; Strong et al., 2011). The Lake Eyre Basin is a large dust source region in Australia and one of the most active dust producing regions in the world averaging 80 dust entrainment events per year (Bullard & McTainsh, 2003; Strong et al., 2011; Washington et al., 2003). Back trajectory atmospheric modeling has shown that during the austral spring, when dust entrainment from Lake Eyre is highest, air parcels originating over Lake Eyre have the capacity to transport dust as far as New Zealand (McGowan & Clark, 2008; Neff & Bertler, 2015). The Lake Eyre dust emission data set only begins in 1960; however, the data set indicates higher than average emission rates from the start of the record through the 1960s and early 1970s. Dust emission rates from the Lake Eyre basin increase again in the early 2000s during a particularly strong drought in Australia (DustWatch, 2015; McTainsh et al., 2007; Speer, 2013); however, meteorological data indicate that dust emissions from the Australian continent were approximately 4.6 times higher during the 1937–1946 drought than the early 2000s drought (O'Loingsigh et al., 2015). This pattern of high dust emission from the 1930s through the 1970s from various regions of the Australian continent is consistent with our reconstructed dust records from New Zealand lakes.

Dust emissions in semiarid and arid regions are tied to aridity, wind speeds, and anthropogenic disturbance (Field et al., 2009). Both short- and long-term variation in Australian dust emissions have been previously

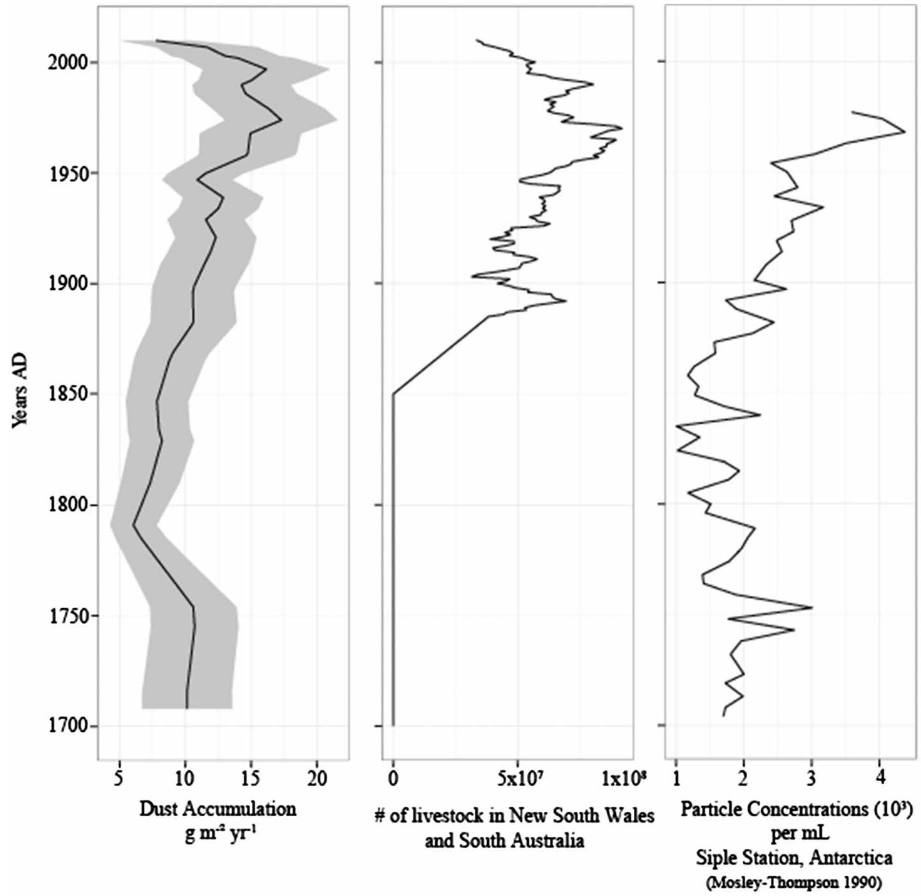

Figure 8. Dust accumulation chronologies as a composite of the two lake records from 1700 AD. Also shown is the number of livestock on the Australian continent between 1885 and 2011. Data includes sheep, horse, and cattle population statistics from New South Wales and South Australia.

linked to variations in Pacific climate variability that affect rainfall intensities over the Australian continent (Marx et al., 2011; Speer, 2013). Dust deposition records presented here indicated a twentieth century increase in dust deposition relative to nearly 2,000 years of sediment record. This increase occurs despite considerable climatic variation during the twentieth century and is strongly suggestive of an underlying shift in continental disturbances. Though several droughts punctuated the twentieth century, including the Federation Drought (1895–1900), the World War II Drought (1937–1945), and the Millennium Drought (2000–2010), long-term climate records do not support drought as the single driver of increased twentieth century dust emissions. Instead, this time period was wetter as compared to the last 1,000 years (Denniston et al., 2015), and average rainfall has increased steadily from 1880 to 1990 AD (Plummer et al., 1999). A distinguishing feature of the twentieth century, however, was the expansion of land area dedicated to pastoralism, agriculture, and mining in Australia. The twentieth century rise in dust concentrations combined with the similarities between historical livestock numbers and the geochemically inferred dust record support hypotheses that land use, specifically agriculture and rangelands, are a significant driver of dust emissions from the semiarid and arid regions in Australia and may exacerbate effects due to natural variability in climate.

A growing body of research is highlighting the potential for far travelled dusts to influences sensitive mountain lake ecosystems (Brahney, Mahowald, et al., 2015; Psenner, 1999). In mountain systems elsewhere in the world, dust particles have been shown to alter water chemistry, nutrient availability, primary production, and community structure (Brahney, Ballantyne, 2015; Jiménez et al., 2018; Reche et al., 2009). Because mountain lake waters are naturally dilute and have limited within catchment capacity to take up deposited nutrients, relatively small deposition rates can induce significant ecological change. For example, in the

Sierra Nevada mountains of Spain dust associated P deposition ranged from 24 to 38 μg P m^{-2} d^{-1}, a small contribution, yet this deposition rate had measurable effects on productivity, inferred from chlorophyll-*a*, bacterial abundance, and plankton diversity (Morales-Baquero et al., 2006; Pulido-Villena et al., 2008). Recently dust deposition has been implicated in the widespread increase in P concentrations in remote lakes and streams across the United States (Stoddard et al., 2016). Both Lake Clara and Adelaide Tarn, and other lakes in the Tasman Mountains, are likely sensitive to atmospheric deposition as their catchments are steep and poorly vegetated. Despite recent declines in dust emissions from the Australian continent, population growth, land use changes, and more frequent and intense droughts (Foley et al., 2005; Hudson, 2011; Trenberth et al., 2014) may lead to future increases in particulate emissions. In light of our results, additional studies on the potential for Australian dusts to influence New Zealand mountain lake ecology are warranted.

5. Conclusions

Two alpine lakes in New Zealand were used to evaluate historical exogenous dust contributions to lake sediments over the last two centuries. Geochemically inferred dust concentration patterns were consistent between the lakes and similar to Australian dust emission inventories through the latter half of the twentieth century as well as Antarctic ice core particulate records that have been verified by model simulations of atmospheric dust transport. These lacustrine records indicate that the twentieth century had unusually high sediment dust concentrations peaking in the latter half of the century, with subsequent declines. Because our recorded shifts in lake sediment dust concentrations vary in concert with land use activities in Australia, we conclude that the most sensible explanation is that twentieth century land use superimposed upon climate cycles has led to enhanced dust emissions and subsequent deposition in New Zealand.

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