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ORIGINAL ARTICLE

Remobilization of trace elements by forest fire in Patagonia, Chile

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Abstract Temporal changes in the amounts of trace elements (As, Co, Cu, Mn, Ni, Pb, and Zn) and their correlations with temporal changes in charcoal abundance in age-dated sediments collected from Lake Thompson in Patagonia, Chile, attest to the substantial pyrogenic remobilization of contaminants that occurred in Patagonia during the mid-1900s. This remobilization was concurrent with the extensive slash and burn period in the region during that period. The changes in concentrations of Co, Cu, and Ni in relation to charcoal abundance in the lacustrine sediments over time were small compared to those of As, Mn, Pb, and Zn. However, the relatively low enrichment factors of all those trace elements, normalized to Fe, indicate that they were predominantly derived from local, natural sources impacted by fires rather than industrial sources. The primarily local source of Pb in the sediments was corroborated by the temporal consistency of its isotopic ratios ($^{206}Pb/^{207}Pb$:$^{208}Pb/^{206}Pb$), which were similar to previously reported values for natural lead in Central and Southern Chile. However, the pyrogenic remobilization of both natural and industrial trace elements by forest fires in Chile and elsewhere is expected to rise as a consequence of climate change, which is projected to increase both the frequency and intensity of forest fires on a global scale.

Keywords Pyrogenic remobilization · Trace elements · Lead isotopic composition · Wildfire

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Introduction

Environmental impacts of contaminant emissions

Multiple human activities release contaminants to the biosphere, and those emissions have dramatically increased since the industrial revolution (Ng and Patterson 1982; Nriagu 1996; Sen and Peucker-Ehrenbrink 2012). Impacts of these emissions on the environment have been extensive, including elevated levels of trace elements in air, surface waters, and terrestrial and lacustrine sediments—both in industrialized and remote uninhabited locations, primarily via aeolian deposition (Nriagu and Pacyna 1988; Boutron et al. 1995; Boyle et al. 2005). For example, emissions from the combustion of leaded gasoline accounted for ~75 % of global Pb aerosols during the second half of the twentieth century (Nriagu 1990; Dunlap et al. 2008), and that industrial lead was dispersed globally (Nriagu and Pacyna 1988; Boutron et al. 1995; Nriagu 1996; Böllhofer and Rosman 2000).

Similar contamination has also been extensively documented in Chile. Air Pb concentrations as high as 1.1 μg/m^3 were measured in Santiago in 1994 (Böllhofer and Rosman 2000), which was over three orders of magnitude higher than the estimated Pb concentration (0.0006 μg/m^3) in a relatively pristine atmosphere (Chow et al. 1972).

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Relatively high concentrations of trace elements (As, Co, Cu, Mn, Ni, Pb, and Zn) were also found in snow collected from regions in the Chilean Andes that were impacted by atmospheric depositions of urban (e.g., leaded gasoline) and industrial (e.g., mining and smelting) emissions compared to their concentrations in snow collected from more pristine sites in the Andes Mountain Range (e.g., Cereceda-Balic et al. 2012). Likewise, Pb and Zn concentrations as high as 129 and 1015 mg/kg, respectively, were measured in topsoil at an industrial site in Talcahuano city (36°S), Chile (Tume et al. 2008). In addition, high levels of As in soil and water have been documented in some parts of Chile, which was the second largest As producer in the world in 2013 (Ferreccio et al. 2000; Hopenhayn-Rich et al. 2000; Edelstein 2014).

Remobilization of trace elements by forest fires

Trace elements sorb to terrestrial sediments, vegetation, and soil organic matter in forests, where they are relatively immobile (Siccama et al. 1980; Friedland et al. 1984; Biswas et al. 2007; Obrist et al. 2008). However, these contaminants are released in more labile forms when vegetation and soil organic matter are burned and volatilized by fire (Young and Jan 1977; Biswas et al. 2007; Wiedinmyer and Friedli 2007). In addition, wildfires increase erosion rates by as much as two orders of magnitude, which subsequently increases the transport rates of remobilized contaminants to water bodies (Cohen et al. 2005; Stein et al. 2012; Warrick et al. 2012). While the impacts of wildfires on Hg influxes to aquatic systems have been relatively well documented (e.g., Kelly et al. 2006), there is much less information on other trace elements (e.g., Pb and Zn) mobilized by wildfires (Erel et al. 1997; Odigie and Flegal 2011, 2014).

Chilean Patagonia is renowned for its relatively pristine environment, extensive freshwater resources, and wildlife (Araneda et al. 2013). However, human activities associated with development in the region began impacting its ecosystems during the last century (Vince 2010; Araneda et al. 2013). Most notably, settlers burned approximately three million hectares of forest between 1930 and 1950 in the Aysén region of Northern Patagonia in Chile, resulting in a loss of ~23 % of its original vegetation (Araneda et al. 2013). The impacts of those deliberate wildfires on ecosystems were, and continue to be, substantial, including the replacement of native species (Coffman et al. 2010; Moreira et al. 2013) and the eutrophication of water bodies (Falcon-Lang 1998)—in addition to the previously noted mobilization of contaminants.

Though wildfires in Central–South Chile over the past century have been largely (~99 %) anthropogenic in origin, both natural and anthropogenic forest fires in that region are projected to be exacerbated by climate change (Gonzalez et al. 2011). For example, temperatures have increased by ~0.4–1.4 °C and annual precipitations have decreased by up to 40 % over the past century in Central–South Chile (Rignot et al. 2003; Glasser et al. 2011; Gonzalez et al. 2011; IPCC 2013).

Study objectives

The extensive forest burning in the Aysén region makes it an ideal environment to study the impacts of wildfires on aquatic ecosystems. Lacustrine sediments are a useful archive of environmental changes, including cycling of environmentally persistent contaminants, in a lake's catchment area (Tierney et al. 2010; Brucker et al. 2011). In addition, Pb isotopic compositions (e.g., $^{206}Pb/^{207}Pb$: $^{208}Pb/^{206}Pb$) may be used as tracers of natural and industrial sources of mobilized Pb in the environment (Ellam 2010; Flegal et al. 2010). Therefore, the objectives of this study were to (1) determine historical trend of trace element concentrations in age-dated sediments of Lake Thompson, Chile, (2) quantify the amount of trace elements (As, Co, Cu, Mn, Ni, Pb, and Zn) released to aquatic ecosystems by wildfires over a temporal scale, and (3) determine the sources of remobilized Pb using its isotopic ratios.

Materials and methods

Study site

Lake Thompson (45°38′26″ S; 71°47′07″ W) is a small, relatively pristine lake located about 20 km southeast of Coyhaique city in Northern Patagonia, Chile (Fig. 1). The lake has a surface area of ~1.18 km^2, maximum depth of ~15 m, and drainage area of ~15.43 km^2 (Araneda et al. 2013).

A sediment core (172 cm) was collected from the deepest part of the lake with an Uwitec gravity corer and subsampled at 1-cm intervals for analyses. Radiocarbon dating was done on a parallel core collected from the lake, and depths of both cores were correlated using magnetic susceptibility and organic content profiles. The dating, organic content measurement, and charcoal analysis are described in detail elsewhere (Bertrand et al. 2012; Araneda et al. 2013). The macrocharcoal particles (>125 μm) in sediment subsamples were disaggregated in hot (70 °C) 10 % KOH solution for 20 min, sieved through a 125-μm mesh, and the retained particles were counted under a stereomicroscope as detailed by Araneda et al. 2013.

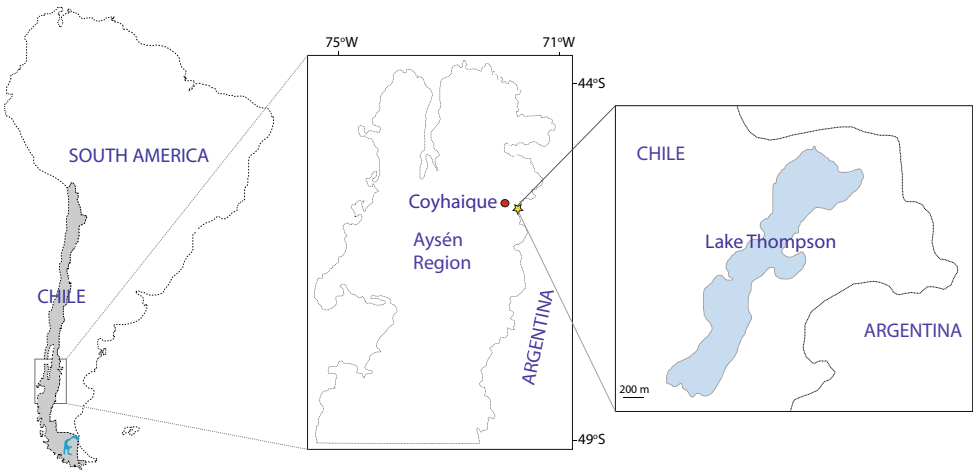

Fig. 1 Map showing the approximate location of the study site, Lake Thompson, in Patagonia, Chile

Analytical methods

All samples were processed with trace metal grade or ultrapure (2× sub-boiling quartz distilled) reagents and high-purity (18.2 MΩ cm) water (Milli-Q) using established trace metal clean techniques (Soto-Jimenez et al. 2006) in a HEPA-filtered (Class 100) trace metal clean laboratory. Concentrations of trace elements (As, Co, Cu, Mn, Ni, Pb, and Zn), concentrations of major elements (Al and Fe), and Pb isotopic compositions (^{204}Pb, ^{206}Pb, ^{207}Pb, and ^{208}Pb) were determined in 41 age-dated sediment samples, including four samples in triplicate. Those samples, triplicates of procedural blanks, triplicates of National Institute of Standards and Technology (NIST) Standard Reference Material (SRM) 1645 (River Sediment), and triplicates of NIST SRM 1646a (Estuarine Sediment) were processed concurrently using established methods (Soto-Jimenez et al. 2006). Approximately 0.3–0.5 g of sediment or SRM was transferred to an acid-cleaned Teflon digestion vial and dried overnight at 65 °C. The sediment was then digested by refluxing in 10 mL of hot (~130 °C) concentrated aqua regia (HNO_3 + HCl, 1:3 v/v) overnight. The digest was dried and redissolved in 10 mL of 1 M ultrapure HNO_3. Concentrations of Co, Cu, Ni, Pb, and Zn of the digests and then their Pb isotopic compositions were determined with a Finnigan ELEMENT 2 high-resolution inductively coupled plasma mass spectrometer (HR ICP-MS). Concentrations of As were determined with a Thermo Scientific XSERIES 2 Quadrupole ICP-MS in collision cell mode. Concentrations of Al, Fe, and Mn of the digests were determined with a PerkinElmer Optima 4300 DV inductively coupled plasma optical emission spectrometer (ICP-OES). Instrumental drifts during analyses were corrected with internal standards (Bi, Ga, In, Rh, Sc, Te, and Y), and instrumental fractionation of lead isotopic composition was corrected with concurrent measurements of NIST SRM 981 (common lead). The detection limit, digestion recovery, and analytical precision for each element are listed in Tables S1, S2, S3, and S4. The digestion recoveries of trace elements in SRM 1645 were higher than those in SRM 1646a, possibly due to differences in their relative chemical compositions.

Results and discussion

The labile (aqua regia digests) concentrations of trace elements (As, Co, Cu, Mn, Ni, Pb, and Zn), concentrations of Al and Fe, and lead isotopic ratios (^{208}Pb/^{207}Pb, ^{206}Pb/^{207}Pb, ^{207}Pb/^{204}Pb, and ^{208}Pb/^{206}Pb) of ^{210}Pb age-dated sediments collected from Lake Thompson, Patagonia, are listed in Table S5. Also listed in Table S5 are the previously reported concentrations of charcoal particles in the sediments (Araneda et al. 2013).

Elemental concentrations

Concentrations (dry weight) of As (1.4–3.7 μg/g), Co (7.0–12 μg/g), Cu (12–17 μg/g), Mn (0.87–2.3 mg/g), Ni (6.1–7.9 μg/g), Pb (4.3–7.1 μg/g), and Zn (36–57 μg/g) in the sediments (Figs. 2 and S1) were generally comparable to reported baseline levels of these elements previously measured in other relatively pristine lacustrine sediments in Chile (Urrutia et al. 2002; Fagel et al. 2010; Palma-Fleming et al. 2012), but were substantially lower (~tenfold) than those measured in lacustrine and river sediments from more industrialized and populated areas in the country (Segura et al. 2006; Fagel et al. 2010). For example, the concentrations of Pb and Zn were as high as 60 μg/g and 1300 μg/g, respectively, in sediments collected from Mapocho River, which runs through and receives anthropogenic inputs from Santiago, Chile (Segura et al. 2006). Concentrations of Al (16.4–26.8 mg/g) and Fe (21.9–46.6 mg/g) in the sediments were also comparable to previously reported

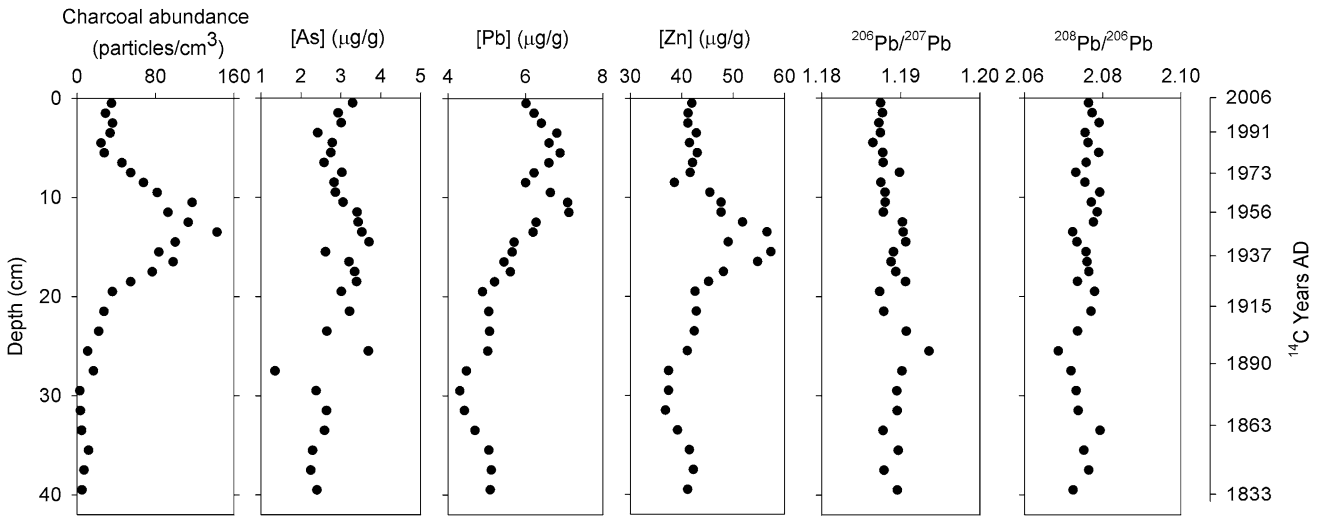

Fig. 2 Charcoal abundance, trace elements (As, Pb, and Zn) concentrations, and lead isotopic ratios ($^{206}Pb/^{207}Pb$ and $^{208}Pb/^{206}Pb$) profiles of sediments collected from Lake Thompson in Patagonia, Chile

levels of these elements in Central and Southern Chile (Urrutia et al. 2002; Chirinos et al. 2005). Therefore, those Al and Fe concentrations are considered to be background or ~natural levels.

Temporal changes in the concentrations of trace elements (Figs. 2 and S1) and their correlations with charcoal abundance (Table S6) in the sediments attest to the impacts of the extensive slash and burn that occurred in Aysén region of Patagonia during the last century. For example, from ~1920 (depth of 19.5 cm) to ~1945 (depth of 13.5 cm), charcoal abundance increased by ~300 %, and during this period, the concentrations of As, Co, Cu, Mn, Ni, Pb, and Zn increased by 38, 13, 13, 40, 11, 27, and 33 %, respectively. These temporal increases were concurrent with the increase in sedimentation rates computed with a parallel core using ^{210}Pb dating (Fig. S3). The substantial concurrent increase in sedimentation rate, which occurred after the massive fires, further attests to the environmental impacts of the historical slash and burn that occurred in Patagonia during the last century (Araneda et al. 2013). The changes in concentrations of Co, Cu, and Ni in relation to charcoal abundance in the lacustrine sediments over time were small (e.g., <20 % from ~1920 to ~1945) compared to those of As, Mn, Pb, and Zn, which increased by more than 20 % over the same period. The concentrations of Zn in the sediments peaked with charcoal abundance (~1940–1960), but the concentrations of Al, Fe, Mn, and Pb in the sediments continued to be relatively elevated for years after that (e.g., levels of Pb appear to be above baseline concentrations in the sediments deposited in the 1990s).

Differences in the rates of transport of remobilized trace elements to the lake could be due to several factors. These include their different affinities to sediments, sensitivities to redox conditions, and solubilities at different pH (Charlatchka and Cambier 2000; Allison and Allison 2005). Differences in the fluxes of the trace elements to Lake Thompson are consistent with their median sediment/water partition coefficients (log K_d, L/kg): Pb (4.2) > Ni (3.1) = Zn (3.1) > Cu (2.7) > Co (2.1) (Allison and Allison 2005). Those differences are also consistent with previous studies that have shown the flux of Pb could be up to two orders of magnitude slower than those of other trace elements in soil, because of its relatively high K_d (e.g., Pang et al. 2004). Correlations of the concentrations of the first-row transition elements, except Zn, with charcoal abundance were low and not statistically significant ($p > 0.05$, $n = 30$, Pearson's correlation), which is attributed to their relatively low affinity to organic matter in acidic solutions (Kraus and Moore 1953).

Pre-1880 levels of some of the elements, including the major elements (Al and Fe), were relatively high—before charcoal abundance started increasing (Figs. 2 and S1), suggesting that other anthropogenic activities (e.g., farming, ranching, and mining) had accelerated the erosion of terrestrial sediments before the intentional use of fires to clear the land. But the concentrations of most of those elements decreased substantially (to their lowest levels) after this period before they started increasing in tandem with the increase in charcoal abundance. The temporal trend of charcoal abundance in the sediments, which peaked during the middle of the previous century,

$$\mathrm{EF} = \frac{\frac{[\mathrm{X}]_{\mathrm{Sample}}}{[\mathrm{Fe}]_{\mathrm{Sample}}}}{\frac{[\mathrm{X}]_{\mathrm{Baseline}}}{[\mathrm{Fe}]_{\mathrm{Baseline}}}}$$

is consistent with the period of slash and burn in the region (Araneda et al. 2013).

Enrichment factors (EFs) of the trace elements were calculated by normalizing the concentrations of trace elements (X) and Fe in each sample to their respective average background concentrations in the sediments (Schiff and Weisberg 1999; Odigie and Flegal 2014). These EFs are considered conservative because the samples were not treated with concentrated hydrofluoric acid, which is required to completely dissolve aluminosilicates (Hornberger et al. 1999; Schiff and Weisberg 1999). The relatively low EFs of the trace elements, which were <5, and their relative consistency over the past century (Fig. S2) indicate that all of them—including Pb—were predominantly derived from local sources. EFs calculated by normalizing trace element concentrations to Al concentrations were similar, indicating little to no effects of redox reactivity of Fe on the EF calculations.

Lead isotopic composition

The predominantly local, natural source of Pb in the sediments was further indicated by their Pb isotopic ratios ($^{206}Pb/^{207}Pb$ vs $^{208}Pb/^{206}Pb$), which have remained relatively constant over the past century and are consistent with previously reported natural ranges of Pb isotopic ratios for Central Chile (Figs. 2 and 3) (e.g., Fagel et al. 2010). The Pb isotopic ratios of these sediments are distinct from those of alkyl lead aerosols measured in Santiago and other cities in Chile. For example, $^{206}Pb/^{207}Pb$ and $^{208}Pb/^{207}Pb$ ratios were ~1.064 and ~2.336, respectively, in Punta Arenas and Puerto Natales in Southern Chile and $^{206}Pb/^{207}Pb$ and $^{208}Pb/^{207}Pb$ ratios were 1.150–1.182 and 2.413–2.436, respectively, in Santiago in the mid-1990s (Böllhofer and Rosman 2000). These spatial variations in Pb isotopic compositions of aerosols reflect the mixing of natural Pb, alkyl Pb, and industrial Pb emissions and the different sources of alkyl Pb used in the region (Fagel et al. 2010). The replacement of leaded gasoline with unleaded gasoline in Chile began in 1993, and leaded gasoline was banned there in 2001 (Pino et al. 2004; Tchernitchin et al. 2005).

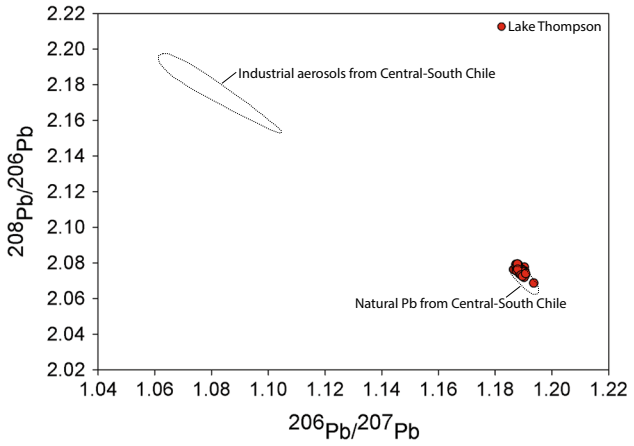

Fig. 3 Lead isotopic ratios ($^{206}Pb/^{207}Pb$: $^{208}Pb/^{206}Pb$) of sediments collected from Lake Thompson in Patagonia, Chile. Also included in the plot are the ranges of natural and industrial lead isotopic ratios of aerosols collected from Central–South Chile (Böllhofer and Rosman 2000; De Vleeschouwer et al. 2008)

Plots of the Pb isotopic ratios indicate that the fraction of anthropogenic Pb in the sediments is small. This relative purity is consistent with the results of an earlier study of sediments in Lago Puyehue, another relatively pristine lake in the Andes in Chile, which determined that anthropogenic Pb accounted for <5 % of the Pb deposited in the lake (Fagel et al. 2010). Consequently, increases in the flux of Pb and—by association—other trace elements (Figs. 2 and S1) in Lake Thompson are predominantly attributed to increased weathering and accelerated rates of erosion during the extensive slash and burn that occurred in the region, rather than temporal increases in industrial emissions over the last century (Fig. S3). Notably, these Pb isotopic ratios show that the increase in Pb concentrations at the middle of last century and then the slow decline are not due to the introduction and subsequent phase out of leaded gasoline in Chile—in spite of the similarity of the temporal variability of the Pb concentration profile in sediments in Lake Thompson to those in sediments of lakes and rivers elsewhere that corresponded with the use of leaded gasoline during the previous century (e.g., Callender and vanMetre 1997; Renberg et al. 2001; Van Metre and Mahler 2004; Lima et al. 2005).

Potential environmental and health implications

Still, the remobilization of trace elements by forest fires is of increasing concern because those fires are expected to increase in frequency and intensity in many parts of the world, including Chile, as a consequence of climate change (Flannigan et al. 2005; Westerling et al. 2006; Gonzalez et al. 2011). Those emissions will also be more problematic.

Trace element loadings in stormwater runoff from burned areas can be up to two orders of magnitude greater than those in runoff from comparable unburned area (e.g., Stein et al. 2012). The remobilization of trace elements and other potential toxicants may be exacerbated by ongoing and planned developments in Southern Chile and other relatively pristine areas—which are expected to cause substantial increases in sediment erosion and deforestation (Vince 2010).

Some of these trace elements are toxic—even at low levels. For example, health impairments continue to be

reported at sublethal blood Pb (PbB) levels that were previously considered harmless (Wheeler and Brown 2013). Although the amounts of trace elements mobilized in Patagonia by wildfire are relatively small compared to those mobilized in more industrialized regions (e.g., Kristensen et al. 2014; Odigie and Flegal 2014), increased remobilization of Pb by wildfire could become problematic.

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