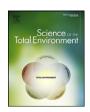
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Historical records of mercury in southern latitudes over 1600 years: Lake Futalaufquen, Northern Patagonia



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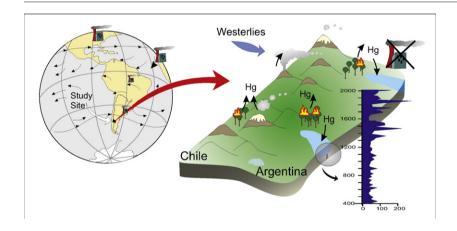
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HIGHLIGHTS

Mercury is a natural element globally distributed affecting remote areas.

- A short core from L. Futalaufquen, Patagonia, was analyzed for Hg time evolution.
- High natural baselines and relative low post-industrial increase were measured.
- Natural/anthropogenic fires and volcanism were the main sources of Hg peaks.
- Lakes from southern latitudes provide valuable records for atmospheric processes

GRAPHICAL ABSTRACT



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ABSTRACT

Mercury is released to the environment from natural and anthropogenic sources, and through atmospheric transport is distributed globally. Lake Futalaufquen (42.8°S) is an oligotrophic lake located in Los Alerces National Park (Northern Patagonia), providing a remote and unpolluted study system. A lacustrine sedimentary sequence revealed 1600 years of Hg deposition, identifying natural baselines and marked peaks not correlated with longrange atmospheric transport. Organic matter and catchment erosion were discarded as Hg drivers. Natural background, pre-1300 CE Hg concentrations, ranged between 27 and 47 ng g $^{-1}$ (accumulation rates from 8 to 15 µg m $^{-2}$ y $^{-1}$). From 1300 CE on, the Hg background profile did not follow the generally increasing Hg pattern observed in both Southern and Northern Hemisphere since pre-industrial times. It was not until the last century that a 1.6-fold increase is observed in the Hg accumulation rate, considered among the lowest increments in southern South America. Noteworthy local/regional sources of Hg for this area, along with global transport, are forest fires and volcanic activity. Between approx. 1340 and 1510 CE, sharp increase in Hg concentration and accumulation rate (up to 204 ng g $^{-1}$ and 51 µg m $^{-2}$ y $^{-1}$, respectively) were clearly associated with extended fire episodes. Furthermore, high Hg peaks during the last 300 years were associated with volcanic eruptions in

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northernmost Patagonia together with fairly irregular fire episodes, caused by anthropogenic burning by settling population in the Andes.

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1. Introduction

Mercury (Hg) is a heavy metal dispersed in the atmosphere by natural processes (volcanoes, geothermal vents, forest fires, geologically Hg-enriched soil), as well as by human activities (mining, coal-fired power plants, landfills, sewage sludge treatment plants) (Bagnato et al., 2014; Driscoll et al., 2013; Lindqvist et al., 1991; Nriagu, 1989; Schroeder and Munthe, 1998). As global pollutant, Hg travels long distances from sources via atmospheric transport, impacting terrestrial, lacustrine, and marine ecosystems (Fitzgerald et al., 1998; Pirrone and Mahaffey, 2005), even in remote regions like the Arctic and Antarctic (Durnford et al., 2010; Sun et al., 2006).

Mercury deposition from the atmosphere increase the global pool of Hg in surface sinks. This depends on deposition mechanisms and meteorological conditions. After deposition, anthropogenic or natural processes result in sequestration or emission back to the atmosphere. Continental water bodies and aquatic sediments are reservoirs of deposited Hg, recording Hg impact in the watersheds (Driscoll et al., 2013; Sprovieri et al., 2010). Thereupon, lake and peat bog sediments can be used as archives of Hg accumulation, integrating local, regional, and global environmental signals (Fitzgerald et al., 1998). Mercury determinations in lake, peat bog, and ice sequences have shown significant changes in prehistoric deposition rates due to climate forcing, i.e. changes in precipitation, air circulation patterns and temperature regimens, extended fires, and volcanic activity (Hermanns and Biester, 2013a; Lacerda et al., 1999; Martínez-Cortizas et al., 1999; Roos-Barraclough et al., 2002). In historical times, increase in Hg deposition with respect to prehistoric rates has been reported associated with anthropogenic activities (Allan et al., 2013; Muir et al., 2009; Perry et al., 2005; among others), with a significant change in the temporal trend due to industrialization (Fitzgerald et al., 1998; Lacerda et al., 1999).

Furthermore, aquatic systems are considered highly sensitive to Hg (Grigal, 2002). In Northern Patagonia, where no relevant Hg releases associated with industrial activities or mining were identified, high Hg concentrations were reported during the last decade. Lichens and mussels were studied as air and water bioindicators; the Hg concentrations measured were compatible with those at locations exposed to moderate contamination (Ribeiro Guevara et al., 2004a, 2004b). High Hg concentrations were measured in lacustrine biota, notably high in plankton, reaching 240 μ g g⁻¹ dry weight (DW) in Lake Moreno and Lake Nahuel Huapi (Rizzo et al., 2014), and in fish muscle, up to 2–3 μ g g⁻¹ DW in different lakes including Lake Futalaufquen (Rizzo et al., 2011). Also, high Hg concentrations, up to $3 \mu g g^{-1}$, were measured in Northern Patagonia lake sediments, far above the values determined in noncontaminated fresh water sediments, together with very high Hg fluxes corresponding to 14th and 18-19th century's deposition (Ribeiro Guevara et al., 2010).

Mercury records in lacustrine sedimentary sequences can be associated with the impact of Hg sources in the watershed, both natural and anthropic at local, regional, or global scale. The study of Hg concentration profiles in dated sedimentary sequences provides valuable information on the Hg sources to aquatic ecosystems, and on environmental events and processes that are associated with Hg concentration variations along the profile. In the present work, Hg concentrations were studied in a 1600 years sedimentary sequence extracted from Lake Futalaufquen, close to the Southern Volcanic Zone, located in a remote, protected area with low anthropogenic pressure but high Hg levels in biota. Regional volcanic activity is tested as a Hg source, as well as fires and global transport.

2. Study site

Lake Futalaufquen (42°49′ S, 71°43′ W) is an oligotrophic system, with 168 m max. depth, located in Los Alerces National Park (LANP) at 518 m above sea level (Fig. 1), in the Northern Patagonia Andean Range (40°15′ to 41°25′ S, 71° to 72°45′ W). Lake Futalaufquen extends over 44.6 km² of old fluvial valleys deepened by glacial erosion, collecting waters from a 2920 km² catchment area characterized by high mountains and several lakes. The main tributary is the river Arrayanes, which drains lakes Rivadavia and Menéndez (Fig. 1). Vegetation is dominated by pure or mixed stands of conifer (*Austrocedrus chilensis*), evergreen (*Nothofagus dombeyi*), and deciduous species (*Nothofagus pumilio* and *Nothofagus antarctica*). Shrub lands dominate between 1000 and 1200 m a.s.l. (Pizzolón, 1995; Vila and Borrelli, 2011).

The Andean Range is located in the Southern Volcanic Zone (SVZ, 33°–46° S) of the Andes, including several active volcanic systems (Stern, 2004). Volcanoes close to the study zone are Huequi, Chaitén, Michinmahuida, and Corcovado (Fig. 1), with partial records available on historical volcanic activity (González-Ferrán, 1995; Petit-Breuilh Sepúlveda, 2004). As it was observed during the last explosive eruption of volcano Chaitén in 2008, volcanic events in the nearby SVZ have frequently affected the Patagonian territory, and the study site in particular (Alfano et al., 2011; Daga et al., 2016).

Climate is cool–temperate humid, with austral fall–winter precipitation, and 8 °C mean annual temperature. The Andes mountains position generate a strong west–east gradient of precipitation across the region, with more than 2000 mm y^{-1} at the eastern base of the Andes decreasing exponentially to under 200 mm y^{-1} on the steppe (Paruelo et al., 1998; Roig and Villalba, 2008).

LANP is a 2600 km² protected area, where anthropogenic activity is limited due to low population density and seasonal tourism. Thus there is little anthropic disturbance of the lacustrine sedimentary record. The first inhabitants in the area were settled around 3000 years ago in the river Desaguadero valley. They were huntergatherer (http://www.parkswatch.org/parkprofile.php). Nowadays about 2000 people live in the basin, with significant livestock activity in the headwaters. Seasonal tourism is important in the Park (Pizzolón, 1995).

3. Materials and methods

3.1. Coring and sampling

A short sediment core of 79 cm length (5.7 cm diameter) was extracted from Lake Futalaufquen (Fig. 1) with a messenger-activated gravity type corer. The sampling site (42°49′ S, 71°43′ W) was selected in a flat area of the lake bottom, determined after echo sounder survey, at 90 m depth. The sediment core was opened and sectioned into 1 cm slices after visual inspection, or following natural boundaries. Each sediment segment was immediately frozen and then freeze-dried until constant weight was achieved. Tephra layers were visually identified by the colour and the grain size, and confirmed after binocular magnifying glass examination. Five tephra layers were identified in the sedimentary sequence (Daga et al., 2016) (Fig. 1). Physical properties such as water content and dry bulk density (DD) were calculated from weight difference between wet and freeze-dried subsamples (Daga et al., 2016).

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