Environmental Pollution 231 (2017) 262–270

Contents lists available at ScienceDirect

Environmental Pollution

journal homepage: www.elsevier.com/locate/envpol

Mercury contamination level and speciation inventory in Lakes Titicaca & Uru-Uru (Bolivia): Current status and future trends☆

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ARTICLE INFO

Article history:
Received 16 May 2017
Received in revised form
27 July 2017
Accepted 2 August 2017

Keywords:
Mercury
Water
Sediment
Biota
Titicaca

ABSTRACT

Aquatic ecosystems of the Bolivian Altiplano (~3800 m a.s.l.) are characterized by extreme hydro-climatic constrains (e.g., high UV-radiations and low oxygen) and are under the pressure of increasing anthropogenic activities, unregulated mining, agricultural and urban development. We report here a complete inventory of mercury (Hg) levels and speciation in the water column, atmosphere, sediment and key sentinel organisms (i.e., plankton, fish and birds) of two endorheic Lakes of the same watershed differing with respect to their size, eutrophication and contamination levels. Total Hg (THg) and monomethylmercury (MMHg) concentrations in filtered water and sediment of Lake Titicaca are in the lowest range of reported levels in other large lakes worldwide. Downstream, Hg levels are 3–10 times higher in the shallow eutrophic Lake Uru-Uru than in Lake Titicaca due to high Hg inputs from the surrounding mining region. High percentages of MMHg were found in the filtered and unfiltered water rising up from <1 to ~50% THg from the oligo/hetero-trophic Lake Titicaca to the eutrophic Lake Uru-Uru. Such high % MMHg is explained by a high *in situ* MMHg production in relation to the sulfate rich substrate, the low oxygen levels of the water column, and the stabilization of MMHg due to abundant ligands present in these alkaline waters. Differences in MMHg concentrations in water and sediments compartments between Lake Titicaca and Uru-Uru were found to mirror the offset in MMHg levels that also exist in their respective food webs. This suggests that *in situ* MMHg baseline production is likely the main factor

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☆ This paper has been recommended for acceptance by Prof. W. Wen-Xiong.
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http://dx.doi.org/10.1016/j.envpol.2017.08.009

controlling MMHg levels in fish species consumed by the local population. Finally, the increase of anthropogenic pressure in Lake Titicaca may probably enhance eutrophication processes which favor MMHg production and thus accumulation in water and biota.

1. Introduction

The Bolivian Altiplano is one of the largest high plateaus in the world containing two large lakes, Lake Titicaca in the north part and Lakes Uru-Uru/Poopó in the central part of an endorheic system consisting in the Titicaca-Desaguadero-Poopó-Coipasa Salt Lake (Revollo, 2001; Delclaux et al., 2007). Lake Titicaca is the most important water resource of the Andean Altiplano, a major source of fish for ~3 million people and the largest navigable water body in the world lying at an altitude of 3809 m above sea level (a.s.l.). Its ecological functioning and limnology have been widely investigated between 1980 and 2000 (Collot et al., 1983; Pourchet et al., 1994; Mourguiart et al., 1998; Dejoux, 1992). Nowadays, the ecological equilibrium of the region is disturbed by a recent but very intensive urban demography and the intensification of mining activities, fisheries and agriculture around the Lake. For example, Puno Bay, on the Peruvian side of Lake Titicaca, has been identified as a contaminated area by urban and industrial effluents since the 1980's (NorthCote et al., 1989). The most preoccupying issue on the Bolivian side of the Lake (Lago Menor – southern basin of the lake) is the extremely rapid development of El Alto city, which population increased from 95,000 inhabitants in 1976 to around 1.2 million according to the last census (Mazurek, 2012) with minimal land planning. Wastewater from El Alto city, its facilities, manufactures and small scale industries, are discharged in the Katari river, which flows to the Lago Menor with less than 50% of water treated (Chudnoff, 2009). Among the emitted pollutants (e.g., nutrients, traces metals and organic contaminants) (Duwig et al., 2014; Archundia et al., 2017), mercury (Hg) contamination is one of the preoccupying issues in this fragile ecosystem. So far, only three studies focused on Hg contamination in the coastal areas of the great Lake Titicaca (Gammons et al., 2006; Molina and Point, 2014; Monroy et al., 2014) revealing high Hg concentrations in all fish species collected in the northern part of the Lago Mayor, particularly at the Mouth of the Ramis River (up to 1840 ng g^{-1} THg, dw in Orestias agassii) where intense gold mining activities are documented. Hg levels in fish muscle from other coastal areas of the Lago Mayor where found below regulatory health guidelines. No data exists yet in Lago Menor which is considered as the most productive area and is also impacted by anthropogenic inputs.

Downstream Lake Titicaca, Lake Uru-Uru, a man-made reservoir, combines diverse metal and organic pollutions since it receives important discharges from urban and mining activities. Recently, several studies evaluating Hg contamination in Lake Uru-Uru have highlighted high concentrations and percentages of toxic MMHg in the water column and biota (Molina, 2015; Alanoca et al., 2016a, 2016b; Lanza et al., 2016).

In this paper, we report for the first time, a complete inventory of Hg levels and speciation, including inorganic Hg(II), elemental Hg (Hg^0) and mono-methylmercury (MMHg), from samples collected between 2010 and 2016 in the different compartments of Lake Titicaca and Uru-Uru. Hg was measured in the atmosphere (total gaseous Hg - TGM), atmospheric fallouts (i.e., filtered and particulate Hg), water column (i.e., filtered, particulate and dissolved gaseous Hg), sediment (i.e., porewater and solid particles) and biota (i.e., plankton, fish and birds). Concentrations of Hg species are compared with previous studies performed in large or elevated lakes. Biogeochemical and anthropogenic factors influencing the sources, distribution and speciation of Hg in the different limnological compartments and selected sentinel organisms are discussed. Based on these discussions and the observed spatial gradients in the ecosystem, we finally propose scenarii of possible future trends in Hg species levels and their impacts on this ecosystem.

2. Material and methods

2.1. General settings

2.1.1. Lake Titicaca

Lake Titicaca (Fig. 1a) comprises of two nearly separate basins: the great lake named “Lago Mayor” or “Lago Chucuito” (7131 km^2; mean depth = 100 m; max depth = 285 m) and the smaller lake named “Lago Menor” or “Lago Huiñaimarca” (1428 km^2; mean depth = 9 m; max depth Chua trough = 40 m) (Dejoux, 1992). The two basins are connected by the Strait of Tiquina and a single outlet for the lake, the Río Desaguadero, which drains out the southern end of Lago Menor to the central Altiplano (i.e., to Lakes Uru-Uru and Poopó) (Cross et al., 2000; Dejoux, 1992). Because of its geographical location and high altitude (3809 m a.s.l.), Lake Titicaca is subject to the tropical zone climate (rainy season concentrated between December and March) and its hydrological regime is dominated by evaporation (~95%) while rivers outflow represents ~ 5%. The resulting Lake water is alkaline with a salinity close to 1 g L^{-1} (Dejoux, 1992). Sediments of Lake Titicaca are covered with Totoras (*Schoenoplectus Californicus*) in the inner margin (0–2 m depth) and by macrophytes (mostly *Characeae* spp.) in the photic zones (i.e.,~15 m depth) with maximum development between 4.5 and 7.5 m. Macrophytes constitute more than 60% of the total biomass in Lago Menor (Dejoux, 1992) and colonize a third of its bottom (~436 km^2) between 2 and 15 m in depth (Collot et al., 1983). Major sources of anthropogenic Hg, the gold mining centers, are identified at the mouth of the Ramis and Suchez rivers (Fig. 1a). Two other urban wastewater sources to the lake are located in the Puno Bay (Lago Mayor) and Cohana Bay (Lago Menor) originating from Puno and El Alto cities, respectively (Fig. 1a).

2.1.2. Lake Uru Uru

Lake Uru Uru is located in the central part of the Bolivian Altiplano region (Fig. 1b), south to Oruro city where numerous mining (e.g., Au, Ag and Sn) and smelting activities are concentrated. This shallow aquatic ecosystem is a man-made reservoir supplied mainly by the Rio Desaguadero waters. The Lake surface and depth vary between 120 and 350 km^2 and 0.25–1 m, respectively, as function of the seasons. Fishing activities are practiced year round and are an important protein and financial resource for local communities. A strong contrast exists between the Northern and Southern parts of the lake, with higher density of sedge (*Schoenoplectus totora*), grass (*Ruppia* spp.) and algae (Characeae) in the southern part (Tapia and Audry, 2013). Previous investigation (Alanoca et al., 2016a) studying 24 h biogeochemical cycles demonstrated that diurnal variability (e.g., temperature, oxygen) in