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Effects of historical and modern mining on mercury deposition in southeastern Peru

Samuel A. Beal^{1,*}, Brian P. Jackson¹, Meredith A. Kelly¹, Justin S. Stroup¹, and Joshua D. Landis¹

¹Department of Earth Sciences, Dartmouth College, Hanover, NH, USA

Abstract

Both modern anthropogenic emissions of mercury (Hg), primarily from artisanal and small-scale gold mining (ASGM), and preindustrial anthropogenic emissions from mining are thought to have a large impact on present-day atmospheric Hg deposition. We study the spatial distribution of Hg and its depositional history over the past ~400 years in sediment cores from lakes located regionally proximal (~90–150 km) to the largest ASGM in Peru and distal (>400 km) to major preindustrial mining centers. Total Hg concentrations in surface sediments from fourteen lakes are typical of remote regions (10–115 ng g⁻¹). Hg fluxes in cores from four lakes demonstrate preindustrial Hg deposition in southeastern Peru was spatially variable and at least an order of magnitude lower than previously reported fluxes in lakes located closer to mining centers. Average modern (A.D. 2000–2011) Hg fluxes in these cores are 3.4–6.9 μg m⁻² a⁻¹, compared to average preindustrial (A.D. 1800–1850) fluxes of 0.8–2.5 μg m⁻² a⁻¹. Modern Hg fluxes determined from the four lakes are on average 3.3 (±1.5) times greater than their preindustrial fluxes, similar to those determined in other remote lakes around the world. This agreement suggests that Hg emissions from ASGM are likely not significantly deposited in nearby downwind regions.

Introduction

Anthropogenic emissions of mercury (Hg) to the atmosphere have more than doubled over the past 60 years, rising rapidly in the past 10 years.^{1, 2} The single largest source of Hg to the environment is currently artisanal and small-scale gold mining (ASGM).³ ASGM uses elemental Hg (Hg⁰) to amalgamate gold from alluvial ores, followed by heating of the amalgam to volatilize Hg and recover gold. In addition to recent emissions, past anthropogenic Hg emissions have been shown to have a persistent effect on Hg in the environment, comprising 60% of present-day Hg deposition.⁴ The majority of past anthropogenic Hg emissions are estimated to have been derived from gold and silver mining during preindustrial time in South and Central America (A.D. 1570–1800) and subsequently globally, during the gold rush (A.D. 1860–1920).¹ The fate of Hg released from both ASGM and historical mining is dependent on the speciation of emissions—Hg⁰, Hg²⁺, or particulate-bound Hg—and the environmental factors governing re-mobilization and volatilization. Because Hg can be transported in the atmosphere globally as gaseous Hg⁰, it is critical to understand the fate and quantity of Hg released by ASGM and historical mining.

*Corresponding Author Information: samuel.a.beal@dartmouth.edu, Phone: 603-646-0921, Fax: 603-646-3922.

Supporting Information Available

Site-specific physical and chemical data; correlations between physical and chemical properties; ²¹⁰Pb activity profiles; cryptotephra identification; and metal time series plots and data. This information is available free of charge via the Internet at <http://pubs.acs.org/>.

ASGM occurs throughout most of South America, and the country of Peru is estimated to consume ~30 tonnes of Hg for ASGM per year during recent time.⁵ The department of Madre de Dios in the Amazon of southeastern Peru (Figure 1a) accounts for 70% of Peru's artisanal gold production, with Huepetuhe being the region's largest mine.⁶ Activity at the Huepetuhe mine has increased rapidly with the rising price of gold in the 21st century,⁷ yet the fate of its Hg emissions, like those from most ASGM sites, remains largely unknown.⁵

Peru has been a center for Hg use since at least A.D. 1570 when Hg amalgamation for silver extraction was introduced to the region.⁸ Cinnabar (HgS) was mined and smelted near Huancavelica, central Peru, producing liquid Hg that was then sent to silver mines throughout the Andes, most notably in Potosí, Bolivia.⁹ Although recent models of the global Hg cycle include estimates of Hg emissions from preindustrial mining in South America,^{1, 4} thus far there is only strong evidence for contamination of soils and sediments near these mining sites,^{10–12} with limited evidence of regional atmospheric transport in cores from two lakes in Peru and the Galápagos¹³ and no evidence for global atmospheric transport.¹⁴

This study uses lake sediments, which are reliable archives of Hg,¹⁵ for the following objectives: 1) to assess total Hg concentrations in surface sediments across a broad region of southeastern Peru; 2) to reconstruct Hg fluxes over the past ~400 years and examine their relation to historical mining; and 3) to compare modern Hg fluxes and flux ratios to those in other remote regions and determine the magnitude of recent atmospheric Hg deposition in southeastern Peru.

Methods

Study Region

Sediment cores were collected from fifteen lakes in southeastern Peru spanning two degrees of latitude (13–15° S), two degrees of longitude (70–72° W), and nearly two kilometers of elevation (3100–4900 m a.s.l.) (Figure 1). Underlying bedrock type ranges from Paleozoic sedimentary and Mesozoic-Cenozoic intrusives in the east of the study region to Carboniferous-Cretaceous sedimentary and Cretaceous-Tertiary volcanics in the west, mainly of felsic-intermediate composition. Typical of the Peruvian Puna, vegetation consists primarily of bunchgrasses, shrubs, and, in poorly-drained areas, sedges, rushes, and cushion plants. Generally, precipitation in this region is sourced from the Amazon Basin in easterly mid-upper troposphere flows during austral summer, whereas austral winter is characterized by little to no precipitation and variable air trajectories.¹⁶

Core Collection

Cores were collected in May and June of 2011 using a gravity corer that preserves the sediment-water interface. Coring locations relative to the lake center and water depths varied (see Supporting Information Table S1), but attempts were made to sample near the lake center and away from inflows. Immediately after collection in the field, cores were extruded in 1 cm intervals and transferred to Whirlpak bags using a stainless steel scraper. Of the fifteen lakes, full cores were retained from four: PLS5, PLS8, PLS12, and YC1. These designations are used for convenience in this study, although some of the lakes have formal names (Table S1). PLS5, PLS8, and YC1 are small (0.04–0.09 km²) headwater lakes with small catchment areas (0.1–0.8 km²), whereas PLS12 is a large (9 km²) open-basin lake with a large, well-developed catchment. Although YC1 is located near the margin of Quelccaya Ice Cap, it is isolated from the input of glacial meltwater.¹⁷ Only the upper two centimeters of sediment (0–1 and 1–2 cm intervals) were collected from the other eleven lakes.

Dating and Chronologies

Lead-210 dating, radiocarbon (^{14}C) dating, and event stratigraphy were used to determine chronologies for the four sediment cores. Freeze-dried samples were homogenized in an agate mortar and pestle and measured for short-lived radionuclides in a Canberra Ge Well Detector. Calibration and data analysis for ^{210}Pb followed Landis et al. (2012).¹⁸ Supported ^{210}Pb was determined by the asymptotic behavior of ^{210}Pb activity with depth (Figure S1). Lead-210 ages were calculated using the Constant Rate of Supply model.¹⁹ Depth-age models for cores from PLS5, PLS8, and YC1 (Figure 2) are extended beyond the ^{210}Pb dating horizon by linear interpolation between the basal ^{210}Pb age and either a cryptotephra layer (PLS5 and PLS8) or a ^{14}C age of an aquatic macrofossil (YC1; Table S2). The cryptotephra layer, likely from the A.D. 1600 eruption of Huaynaputina in southwestern Peru, was identified by bulk geochemistry and SEM images of acid-extracted sediment (Figures S2–S4). The age model for PLS12 was not extended beyond the ^{210}Pb dating horizon because a cryptotephra was not apparent in its bulk geochemistry and macrofossils for ^{14}C dating could not be reliably extracted from the sectioned sediment.

Geochemical Analysis

Freeze-dried and homogenized sediments were analyzed for total Hg using a Milestone DMA-80 direct mercury analyzer. Total Hg concentrations are expressed as nanograms of Hg per gram of dry sediment (ng g^{-1}). Duplicates and one of the standard reference materials (SRMs, i.e. IAEA-SL-1, STSD-1, or NIST 1547) were run every ten samples. Recoveries of total Hg in each of the SRMs were within the published 95% confidence intervals (Table S3). Sediments were also digested by strong acid (3:1 $\text{HNO}_3\text{:HCl}$) in open microwave vessels at 90 °C and analyzed for major and trace elements by quadrupole ICP-MS (Agilent 7500, 7700). Loss on ignition at 550 °C (LOI_{550}) was used to estimate organic matter.

Hg Flux Calculations

Hg fluxes were calculated as the product of Hg concentration and sedimentation rate. Sedimentation rate was calculated from accumulation rate (cm a^{-1}), derived from the slope of the depth-age model, and bulk density for a given sediment interval. Hg flux ratios were calculated for each sediment interval as Hg flux in that interval normalized to the average Hg flux in its core for the period A.D. 1800–1850. This period is often used to represent preindustrial conditions, permitting comparison of this work to other studies, but may not represent natural conditions.²⁰ The preindustrial period for Hg flux ratios in PLS12 was defined as A.D. 1875–1900 because of its basal age of A.D. 1875. This may result in an underestimation of flux ratios in this core. A modern flux ratio was calculated for each core as the average of post-A.D. 2000 flux ratios.

Results and Discussion

Surface Sediments

Average total Hg concentrations in surface sediments (0–1 and 1–2 cm intervals) are relatively high ($61\text{--}115 \text{ ng g}^{-1}$) at the higher elevation lakes on the eastern edge of the Andes near the Amazon basin, and are low-to-medium ($10\text{--}60 \text{ ng g}^{-1}$) at lower elevation lakes further from the Amazon (Figure 1). The main exception to this trend is PLS10, located furthest from the Amazon, which has an average Hg concentration of 583 ng g^{-1} . This anomalous Hg concentration, and the lack of similarly elevated concentrations of As, Cd, and Pb (Table S4), indicates a source of Hg within or near the catchment. Overall surface sediment Hg concentrations in fourteen of the fifteen lakes are similar to those in other remote lakes.^{21–25} This finding is important to local ecosystem toxicity as total Hg concentrations in surface sediments are one of the major controls on the formation of

extremely toxic methylmercury.²⁶ Inter-lake variations in surface sediment Hg concentration are likely controlled primarily by organic matter content, evident in the only significant correlation between surface sediment Hg concentrations and LOI₅₅₀ (Figure 3a, Table S5). The association of Hg and organic matter has been demonstrated in numerous studies in various locations and down-cores.^{23, 24, 27}

Down-Core Variations

In the four sediment cores, down-core Hg concentrations (Figure 4) and LOI₅₅₀ are not significantly correlated in PLS5 and PLS8, are significantly positively correlated in PLS12, and are significantly negatively correlated in YC1 (Figure 3b). There is a tendency for Hg concentrations to correlate more strongly to low (<20 %) LOI₅₅₀ sites (Figure 3), suggesting a possible threshold for Hg–organic matter association. Down-core Hg concentrations are not significantly correlated with Ti concentrations in PLS5, PLS8, and YC1 during the preindustrial or the post-industrial periods (Figure S5). Hg and Ti concentrations are significantly correlated only during the preindustrial period in PLS12. From these correlations, it is apparent that down-core variations in Hg are independent of: 1) total organic matter content in at least three of the sediment cores, and 2) lithogenic input represented by Ti and derived from either surface runoff in the catchment or dust input.

Historical Hg Fluxes

Robust depth-age models (Figure 2) for PLS5, PLS8, PLS12, and YC1 permit the reconstruction of historical Hg fluxes. Modern Hg fluxes in these cores are 3.4–6.9 $\mu\text{g m}^{-2} \text{a}^{-1}$, compared to preindustrial Hg fluxes of 0.8–2.5 $\mu\text{g m}^{-2} \text{a}^{-1}$ (Figure 5a). The convergence of preindustrial fluxes to this narrow range in the four cores suggests that these lakes record not only secular changes in Hg deposition but also an approximation of absolute atmospheric Hg deposition rates, without invoking focusing corrections. The preindustrial and modern Hg fluxes are very similar to those from other remote lakes, notably in the very well-characterized lakes (~10 cores per lake) from northern Alaska (modern: 4–8 $\mu\text{g m}^{-2} \text{a}^{-1}$; preindustrial: 1–2),²² suggesting a general coherence of both preindustrial and modern Hg fluxes across hemispheres.

PLS8 has a pronounced short-lived peak in Hg flux beginning around A.D. 1600, and PLS5 has a slight increase in Hg flux from A.D. 1640 to 1850 (Figure 5a), both coincident with the onset of large-scale Hg use for silver amalgamation in the Andes.⁹ Increased Hg fluxes during this period are largely independent of any other change in geochemistry in PLS5 (Figure S6) and in PLS8 (Figure S7). The correspondence of the top of the identified cryptotephra layer with the beginning of the peak in Hg fluxes in PLS8 may indicate a volcanic source. However, because of the lack of an increased Hg flux at this time in the other cores and the non-correlation of Hg fluxes and visible tephra layers from other eruptions in South America,²⁸ it is likely that anthropogenic Hg emissions, rather than volcanism, are responsible for this peak in preindustrial Hg fluxes in PLS8. Increasing Hg fluxes in YC1 from A.D. 1780 to 1910 relative to depressed fluxes in PLS5 and PLS8 during this time indicate a potentially separate source of Hg to YC1. The discrepancy between preindustrial Hg fluxes in these three lakes, located within 70 km of each other, indicates that the distribution of preindustrial Hg emissions was highly variable. The distribution likely depended on the location of the Hg source, the prevailing winds at the time of emission, and the speciation of emissions. Cooke et al. (2013) used Hg isotopes to show the regional transport of Huancavelica-derived Hg to Laguna Negrilla, ~200 km southeast of Huancavelica and ~300 km northwest of PLS5 and PLS8, but they were not able to distinguish whether this Hg was Hg⁰ or particulate-bound.¹³ The muted signal of preindustrial Hg fluxes in PLS5, PLS8, and YC1, and the altogether low preindustrial fluxes relative to other lake cores (average of ~7 $\mu\text{g m}^{-2} \text{a}^{-1}$)¹⁵ and Laguna Negrilla (up to 82 μg

$\text{m}^{-2} \text{a}^{-1}$),¹³ suggest a rapidly decreasing impact of preindustrial mining emissions with distance from Huancavelica. Because of uncertainties in the reconstructions and highly variable wind vectors in this region,¹⁶ further research is needed to establish the spatial distribution of Hg emissions from preindustrial mining.

Subsequent to the convergence of Hg fluxes in PLS5, PLS8, and PLS12 at ~A.D. 1860 to $1.0\text{--}1.6 \mu\text{g m}^{-2} \text{a}^{-1}$, Hg fluxes generally increase into the 21st century (Figure 5a). In PLS5 and PLS8, an initial increase in fluxes of Fe, Cu, As, Cd, and Pb occurred ~A.D. 1950, possibly due to the start of industrial mining in the surrounding area (Figures S6 and S7). The initial peak in these metal fluxes is not reflected in Hg fluxes in PLS5, but may explain the peak in Hg fluxes in PLS8 centered ~A.D. 1980 as a mining dust source that delivered all of these metals to the lake. Similarly, the peak in Hg fluxes in YC1 from ~A.D. 1950 to 1980 is synchronous with increases in all of the other analyzed metals, implying either a source of Hg from mining dust or an increase in lithogenic input to the lake (Figure S8). The late 20th and early 21st century increase in Hg flux in PLS12 is joined by increases in Cu, As, Cd, and Pb fluxes, but not Ti or Fe, suggesting a synchronous anthropogenic source of these metals to this lake (Figure S9).

Modern Flux Ratios

Hg flux ratios show a general unremitting increase over the past 100 years in all of the cores except YC1 (Figure 5b). The mean modern Hg flux ratio determined from the four lake cores is 3.3, with a standard deviation of 1.5. The high standard deviation is in part due to the low modern flux ratio for YC1 (1.4) that is affected by its relatively elevated preindustrial Hg fluxes. Thus, the mean modern flux ratio from these four cores serves as a conservative but likely under-estimate. Although this study's mean modern flux ratio is only slightly greater than those reported from other remote lakes (Table 1), it is well within the typical range reported for lake sediment cores.¹⁵ The lack of an enhanced modern flux ratio in this study indicates that regional ASGM is likely not inordinately affecting Hg deposition in the study region. If Hg from ASGM is not significantly deposited in nearby down-wind regions, then the Hg released by ASGM either remains locally deposited near the mines or directly enters the global Hg pool. Both of these fates likely occur, and understanding the fraction of total Hg released by ASGM to either of these fates is important for constraining the environmental impacts of ASGM. Furthermore, the general agreement of modern flux ratios between the sites in Table 1 suggests that recent anthropogenic Hg has been distributed with some uniformity around the world.

Although commonly used for determining flux ratios, the period A.D. 1800–1850 is likely not representative of natural conditions. Amos et al. (2013) used estimates of preindustrial anthropogenic emissions, primarily from silver mining in South and Central America,^{1, 8} to model the global mercury cycle and determined that the modern atmosphere has 7.5 times more Hg than the natural atmosphere.⁴ Although the depth-age models for PLS5 and PLS8 do not permit Hg reconstruction before A.D. 1600, and for YC1 before A.D. 1460, Hg fluxes in the oldest dated sections of each core are within error of their average A.D. 1800–1850 values and equal to some of the lowest reported Hg fluxes in lake sediments.¹⁵ This finding suggests that modern Hg fluxes to this region are approximately 3 times higher than both preindustrial and natural fluxes. Older sediment cores are needed to confirm accurate rates of natural atmospheric Hg deposition.

Supplementary Material

Refer to Web version on PubMed Central for supplementary material.

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Figure 1.

a) Map of Peru with the department of Madre de Dios highlighted in orange and the study region indicated by the red box. Black arrows represent NCEP/NCAR reanalysis V1 annual average vector wind at 500 mb from A.D. 1948–2012.²⁹ **b)** Enlarged digital elevation model of the study region. Shown are the fifteen studied lakes labeled by their average surface sediment (0–2 cm) total Hg concentration.

Figure 2. Depth-age models and sedimentation rates (SR) for the four lake sediment cores used to calculate historical Hg fluxes.

Figure 3.

a) Average surface sediment (0–2 cm) Hg concentrations in all lakes (excluding PLS10) correlated to LOI₅₅₀. **b)** Down-core Hg concentrations in the four lake sediment cores correlated to LOI₅₅₀. Pearson correlation coefficients and significance levels are denoted for each of the correlations (* indicates $P < 0.05$, ** indicates $P < 0.01$).

Figure 4.

Hg concentrations with depth in the four lake sediment cores. Dashed lines represent the oldest limit of each core's chronology.

Figure 5.

a) Reconstructed Hg fluxes for the period A.D. 1500–2011. **b)** Hg flux ratios for the period A.D. 1800–2011. Error bars represent ± 1 standard deviation.

Table 1

Modern flux ratio means and standard deviations (SD) for this study compared to those from other remote lakes. The modern decade is the average age of the modern flux used to calculate the flux ratio, rounded to the nearest decade.

Site	<u>Modern Flux Ratio</u>		Modern Decade	Reference
	Mean	SD		
Southeastern Peru	3.3	1.5	2010	This Study
Northern Alaska	2.5	1.0	1990	(22)
Western Canada	1.8	0.8	2000	(24)
Uganda	2.8	0.2	2000	(25)
New Zealand	3.0	0.5	2000	(14)
Patagonia	1.9	0.5	2010	(30)