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Short Communication

Past and present mercury flux to a West African crater lake (Lake Bosomtwe/Bosumtwi, Ghana)

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ARTICLE INFO

ABSTRACT

Lake sediment cores have been used to reconstruct mercury deposition patterns in many parts of the world; however, no studies to date have used these methods in West Africa, nor are there any published measurements of mercury deposition to this region. We measured mercury in a ^{210}Pb dated sediment core from a meromictic crater lake in West Africa (Lake Bosomtwe, Ghana). Lake Bosomtwe has a very small catchment area to lake area ratio (1.1) and the sediment mercury profile is expected to reflect past and present atmospheric mercury deposition to the lake. Mercury concentrations in sediments as well as mercury flux to the sediments increased from the mid-1800s to latter half of the 1900s, however there has been a sharp decline in mercury flux to Lake Bosomtwe in recent decades. The recent decline in mercury flux to Lake Bosomtwe's sediments does not appear to be consistent with trends in local, regional or global mercury emissions, and may instead reflect declining global atmospheric mercury concentrations or declining European emissions, highlighting the importance of long-range atmospheric transport of mercury.

1. Introduction

The global release of mercury to the environment has increased greatly as a result of human activities (Pacyna et al., 2006). Several countries, including many in North America and Europe, have implemented regulations to reduce the use and emission of mercury with the aim of lessening human and wildlife exposure to this contaminant. However, many other countries have not yet taken these measures, and although emissions of mercury in Europe and North America have decreased in recent decades, emissions from Asia continue to increase and on a global scale, total mercury emissions are generally increasing (Pacyna et al., 2006; AMAP/UNEP, 2008).

In Africa, biomass burning, coal combustion, artisanal and small-scale gold mining (ASGM) and metal processing are primary sources of mercury emissions (AMAP/UNEP, 2008). Population continues to increase rapidly throughout Africa, and mercury emissions from fuel combustion and biomass-burning are likely to increase. In addition to local deposition, mercury is subject to long-range atmospheric transport, therefore, its continued use and emission pose risks to humans and animals both at the point of use and in remote locations (Jackson, 1997). As a result, regional emission estimates may not accurately predict subsequent regional deposition rates or exposure risks.

Dated sediment cores have been used to reconstruct historical and present fluxes of mercury to lakes in many parts of the world including North America (Kamman and Engstrom, 2002; Engstrom et al., 2007; Muir et al., 2009), Europe (Landers et al., 1998), South America (Cooke et al., 2009), the Tibetan Plateau (Yang et al., 2010a), and East Africa (Campbell et al., 2003; Yang et al., 2010b). Mercury fluxes to lake sediments are dependent on a number of factors, including atmospheric and catchment Hg inputs as well as losses through evasion and outflow. As reviewed in Yang et al. (2010b), there has been a broadly observed increase in Hg accumulation in sediment cores (from remote locations) of approximately 3-fold since pre-industrial times. To date, the few published studies of mercury concentrations in African lake sediment cores have all been based in East Africa on Lake Victoria (Campbell et al., 2003; Ramlal et al., 2003) and in small lakes at high elevations in the Rwenzori Mountains of Uganda (Yang et al., 2010b). Here, we report first data on mercury in a sediment core from Lake Bosomtwe/Bosumtwi (hereafter referred to as Lake Bosomtwe), Ghana, a site in equatorial West Africa (6°30′N, 1°25′W).

1.1. Lake Bosomtwe

Lake Bosomtwe is a meteoric crater lake with a diameter of 8 km and a maximum depth of 78 m (Turner et al., 1996; Table 1). Because of its origin as an impact crater (Koeberl et al., 1997), the lake has a very low catchment area:lake area ratio (1.1), and therefore,

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Table 1
Physicochemical characteristics of Lake Bosomtwe (Ghana). Geographic and limnological data are from Otu (2010) and climatic data are from Shanahan et al. (2008).

Parameter	Value
Latitude	6°30′N
Longitude	1°25′W
Catchment area	54.5 km^2
Lake area	48.6 km^2
Maximum depth	78 m
Mean epilimnetic TP	14.9 ± 8.98 μg L^{-1}
Mean Chl a	10.5 ± 4.8 μg L^{-1}
Mean annual precipitation	1260 mm
Mean air temperature	26 °C

atmospheric deposition of mercury on the lake surface is expected to be responsible for an unusually large proportion of the mercury entering the lake. The lake is meromictic with a permanently anoxic hypolimnion below a depth of 30–35 m and the sediments are finely laminated (Otu, 2010; Shanahan et al., 2008).

Seasonality of precipitation and temperature in the Lake Bosomtwe region is largely driven by the oscillation of the Inter-Tropical Convergence Zone (ITCZ; Asnani, 1993). Annual movement of the ITCZ generally results in a bimodal rainfall pattern for the Lake Bosomtwe region, with peaks in rainfall when the ITCZ is overhead and moist air from the Gulf of Guinea meets dry air from the Sahara. Dry seasons typically occur in July–August when the ITCZ is located to the north of the lake and convective precipitation is reduced due to upwelling of cold water in the Gulf of Guinea, and in December–January when the ITCZ is south of the lake, and dry winds from the Sahara region prevail (Nicholson and Palao, 1993; Puchniak et al., 2009; Shanahan et al., 2008).

Lake Bosomtwe is the only natural lake in the Sahel region, and one of few stratigraphic records available in West Africa for characterizing a large, climatically sensitive region. The annual varves present in Lake Bosomtwe's sediments (Shanahan et al., 2008) indicate limited mixing of sediments (and mobility of mercury) once deposited. Furthermore, due to its remarkably low terrestrial catchment to lake area ratio and its hydrologically closed basin, which prevents washout of Hg deposited to the lake and catchment, the Hg record in Lake Bosomtwe's sediments can provide insight into atmospheric Hg deposition to the region.

Characterizing current and historical mercury deposition in West Africa is also of regional interest due to its history as Africa's "gold coast". This region has long been a center of both artisanal and industrial scale gold mining, and although industrial gold mining in Ghana has primarily used cyanide for gold extraction, artisanal gold mining using Hg has been common, and is currently on the increase in Ghana and West Africa as a whole (Hilson, 2002). Lake Bosomtwe, although endorheic, lies in the Pra River basin, an important area for artisanal gold mining (Donkor et al., 2006a); however, there is no known history of mining within the lake catchment itself, so Hg inputs from regional mining activities would need to be atmospheric.

2. Materials and methods

2.1. Sampling methodology

In May of 2004 a sediment core (81 cm long) was collected from the center of Lake Bosomtwe (at 78 m depth) using a modified Kullenberg corer, and is referred to as core 'Central 1' (Fig. 1). The ends of the core were sealed with mesh, foam and cork and the core was transported intact to the University of Waterloo where it was stored in the dark at 4 °C. In 2005, this core was extruded vertically (Glew, 1988) into 0.5-cm intervals (with the exception of the top core section, which had a depth of 1.5 cm) that were placed into
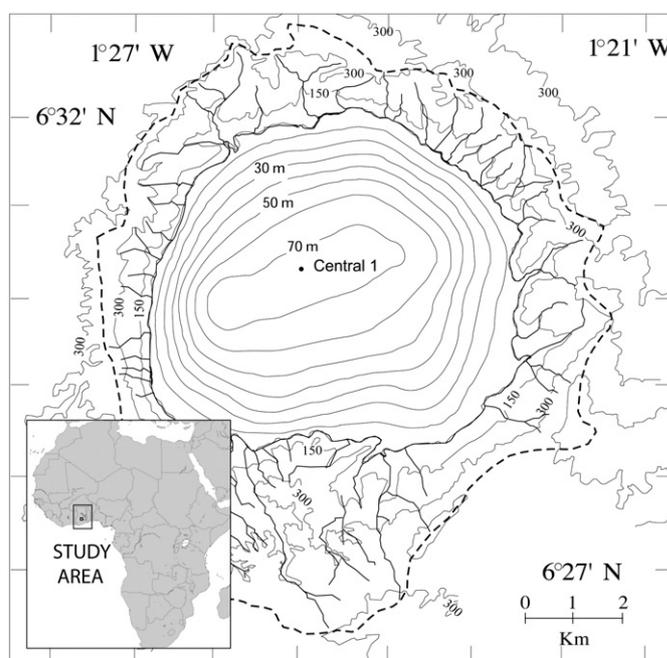

Fig. 1. Map of the Lake Bosomtwe basin showing the catchment topography (meters above mean sea level) and the lake bathymetry (10 m increments). The modern lake level is 99 m above mean sea level. The dashed line denotes the catchment boundary, and the sites from which the Central 1 core was collected are indicated. The inset indicates the location of Lake Bosomtwe in Ghana (West Africa).
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plastic Whirlpak® bags and stored in the dark at 4 °C until further analysis. The core barrel was polycarbonate and core sediments did not come into contact with any metal during coring or processing.

2.2. ^{210}Pb and ^{137}Cs chronology

The sediment core chronology was developed by analyzing the Central 1 core for activities of ^{210}Pb and ^{137}Cs at the Waterloo Environmental Change Research Laboratory (WATER), University of Waterloo. For each sample, a measured mass of freeze-dried sediment was packed into polypropylene tubes, sealed with 1 cm^3 of epoxy resin and allowed to equilibrate for at least 3 weeks before analysis. Activities of the radioisotopes were measured in an Ortec co-axial HPG3 Digital Gamma Ray Spectrometer (Ortec # GWL-120-15). The Constant Rate of Supply (CRS) model (Appleby, 2001), was used to develop a sediment core chronology. Supported ^{210}Pb activity was estimated based on mean ^{210}Pb activity below 18 cm in the core. Peak ^{137}Cs activity, generally expected to coincide with the 1963 peak ^{137}Cs fallout from nuclear testing (Davis et al., 1984), was used to provide an independent dated stratigraphic marker to compare with the ^{210}Pb-based chronology.

2.3. Mercury and organic matter analyses

Subsamples of sediment from the Central 1 core were freeze-dried and homogenized using a mortar and pestle. Total mercury (THg) concentrations in sediment were determined according to EPA Method 7473 (U.S. EPA, 1998), using a DMA-80 direct mercury analyzer. Certified standard reference material (MESS-3) was analyzed in each run to determine between-run variability. The mean coefficient of variation for sample run in duplicate was 1.9 ± 1.8%. Sediment organic matter content was determined through loss-on-ignition, whereby the percent of organic matter was assumed to be the proportion of dried sediment lost following ashing at 550 °C for 1 h.

2.4. Calculations

THg fluxes (in μg m^{-2} y^{-1}) to the sediments at the Central 1 coring site were calculated by multiplying mercury concentrations (in μg g^{-1}) by CRS-modeled dry mass sedimentation rates (in g m^{-2} y^{-1}). Flux ratios between THg flux at a given core depth and calculated flux for 1870 (the deepest core section for which a reliable CRS-modeled sedimentation rate was available) were calculated for all core subsamples.

3. Results and discussion

3.1. Chronology and sedimentation rate

In the Central 1 core, ^{210}Pb activity decreased exponentially with depth, reaching background values at a depth of approximately 18 cm (Fig. 2). In lake sediment profiles, it is expected that ^{137}Cs activities peak in 1963, due to fallout from nuclear bomb testing (Davis et al., 1984). There was good correspondence between the peak in ^{137}Cs activity (assumed to be 1963) and CRS modeled dates (Fig. 2). The unsupported ^{210}Pb inventory for the Central 1 core was 5250 Bq m^{-2}, which corresponds to an unsupported ^{210}Pb flux of 163 Bq m^{-2} y^{-1}. The ^{210}Pb dated portion of the core approximately corresponded to the years 1831–2000, however, due to high associated uncertainty for the deepest core sections (where ^{210}Pb activity approached background levels), we have not presented data from prior to 1870.

Given that Lake Bosomtwe is a closed-basin lake, it is susceptible to lake level changes, which can affect sedimentation rate as well as sediment focusing processes, and as such may affect Hg flux to the sediments. Lake levels in Lake Bosomtwe are thought to have varied by approximately 6 m between 1938 and 2004 (Shanahan et al., 2007) and by approximately 19 m since 1860 (Otu, 2010). However, due to Lake Bosomtwe's steep-sided catchment, changes in lake level do not tend to lead to substantial changes in lake area or catchment area to lake area ratio (Shanahan et al., 2007).

The mean CRS-modeled sedimentation rate for the dated portion of the Central 1 core was 0.0175 g cm^{-2} y^{-1} and ranged from 0.0117 to 0.0254 (Fig. 2). We observed a gradual decrease in sedimentation rate from 1870 to 1950, coinciding with an increase in lake level (Otu, 2010). This is puzzling, since a rainfall driven lake level rise should be associated with increased erosional inputs of inorganic catchment material as well as low ^{210}Pb-activity material from ancient sediments present in the lake catchment. However, sedimentation rates have remained relatively stable since 1960

(0.0165 ± 0.002 g cm^{-2} y^{-1}), a time period that includes both the observed peak as well as the recent decline in Hg flux.

3.2. Mercury concentrations and fluxes

As has been observed in sediment cores from around the world, particularly in North America and Europe, mercury concentrations in the Central 1 core from Lake Bosomtwe have risen appreciably since the mid-1800s (Fig. 3a). However, mercury concentrations peaked in the latter half of the 1900s, and then have declined sharply in recent decades (Fig. 3a). Several recent studies in arctic and subarctic lakes have shown a close relationship between sediment organic matter and mercury concentrations in recent sediments, and have suggested that changes in lake productivity may drive concomitant changes in algal uptake of Hg and deposition to the sediments, making it difficult to infer changes in atmospheric inputs from changes in sediment Hg (Outridge et al., 2007; Stern et al., 2009). In Lake Bosomtwe, % organic matter in the sediments has generally increased over the time period captured by the sediment core (Fig. 3a), likely as a result of increasing primary productivity (Otu, 2010). Although THg concentrations also increase from the mid-1800s until ~1940 (coincident with increases in global Hg emissions), after this point the % organic matter and THg profiles in the Central 1 core diverge (Fig. 3a). This suggests that although we cannot exclude the possibility that an increase in % organic matter was an important contributor to the rise in Hg concentrations prior to ~1940, we are confident that the recent Hg trends in tropical Lake Bosomtwe's sediments are not a reflection of changes in the amount of organic matter, but rather of changing atmospheric inputs.

Calculated mercury flux (based on CRS modeled sedimentation rates) to Lake Bosomtwe's sediments follows a very similar pattern of increase, peak and recent decline as was observed for mercury concentrations, although the increase in flux is much less pronounced (Fig. 3b). The estimated reference (1870), peak (1967), and modern (2000) fluxes of Hg to the sediments of the Central 1 coring site were 13.5, 40.5 and 20.9 μg m^{-2} y^{-1} respectively (Table 2). Declines in Hg concentrations and fluxes have been observed in sediment cores from several lakes in North America and Europe (Kamman and Engstrom, 2002; Engstrom and Swain, 1997; Bindler et al., 2001; Lindeberg et al., 2007). These trends have generally been attributed to the implementation of regulations to reduce the use and emission of mercury that were enacted in the early 1980s. Campbell et al. (2003) observed a peak in Hg concentrations in the sediments of Lake Victoria (East Africa) between 1970 and the early
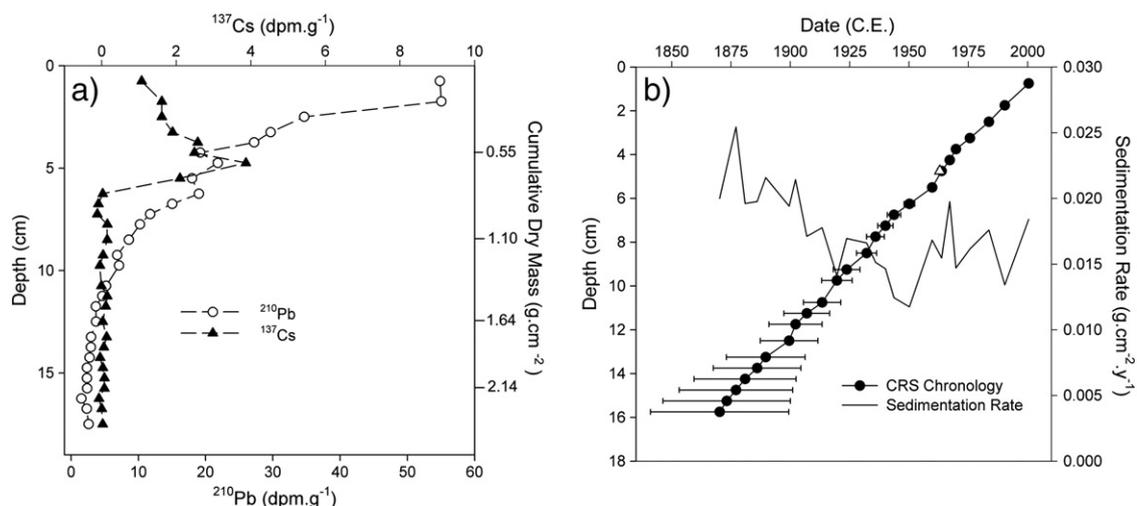

Fig. 2. a. ^{210}Pb (open circles) and ^{137}Cs (black triangles) activity profiles (plotted against depth and cumulative dry mass), and b. CRS chronology (black circles; plotted against depth), 1963 ^{137}Cs activity peak (white triangle; plotted against depth), and CRS modeled sedimentation rates (line graph) for the Central 1 core from Lake Bosomtwe.
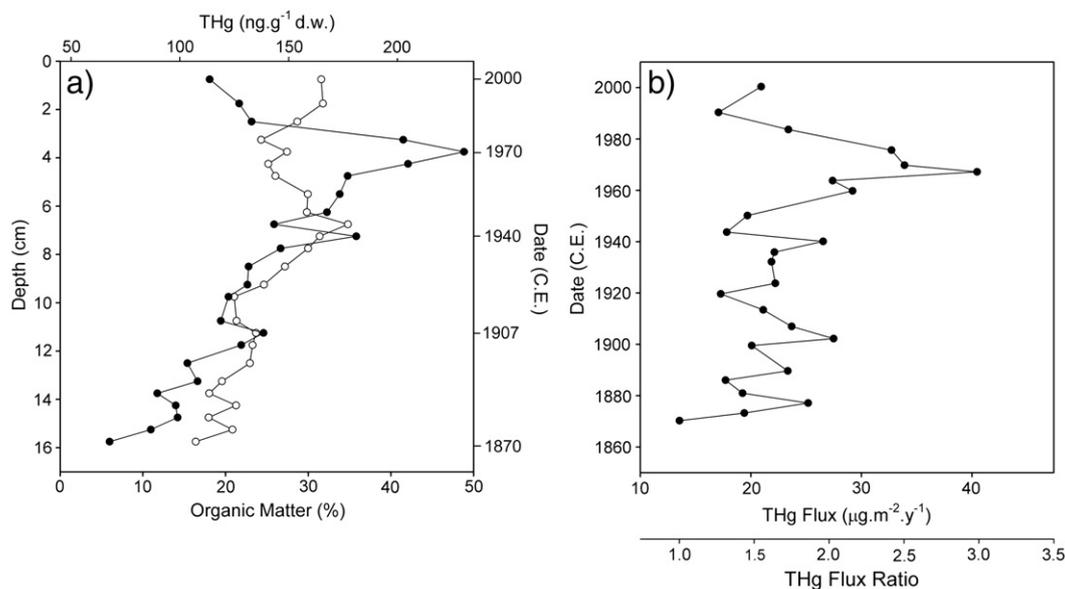
Fig. 3. a. Total mercury concentrations (filled circles) and percent organic matter (open circles) and b. total mercury flux to the sediments and flux ratio (ratio of Hg flux in a given year to 1870 flux) for the Central 1 core from Lake Bosomtwe. CRS modeled dates for the two plots do not align since concentrations and percent organic matter are plotted against sediment depth, while Hg fluxes are plotted against CRS modeled dates.

1980s followed by a decline in Hg concentrations in the most recent sediments. However, given the recent cultural eutrophication of Lake Victoria and the considerable changes that have taken place in the large catchment of this lake (Hecky et al., 2010), the authors could not conclusively attribute this decline to a decrease in atmospheric Hg inputs, since increased sedimentation rates, or dilution of Hg influx by increased organic matter production driven by eutrophication can dilute mercury concentrations in sediments (Campbell et al., 2003). Yang et al. (2010b) observed a recent decline in Hg concentrations in sediments from one of the three Rwenzori mountain lakes included in their study, however, this decrease was attributed to an increase in sedimentation rate apparently resulting from a local watershed disturbance, and was not accompanied by a similar strong decrease in Hg flux to the sediments. Lake Bosomtwe is unique among all published stratigraphic studies of Hg in African lake sediments in that it shows a distinct recent decrease in both Hg concentrations in and flux to the sediments (Fig. 3). Furthermore, because atmospheric deposition dominates the Hg input to Lake Bosomtwe, the recent decline in Hg flux to Lake Bosomtwe's sediments is likely attributable to decreasing atmospheric Hg inputs to the lake.

Although Hg emissions in Europe and North America continue to decline, there is evidence that global mercury emissions are increasing (Pirrone et al., 2009). At a continental scale, rapid population growth throughout sub-Saharan Africa is likely leading to an increase in Hg emissions from industrial activities, coal combustion and biomass burning. In Ghana, mercury emissions from the artisanal and small-scale gold mining (ASGM) sector have likely increased since 1989, when ASGM and use of Hg in these endeavors were legalized, causing a dramatic increase in ASGM activities (Donkor et al., 2006b). Despite the evidence that global, continental and regional Hg emissions are increasing, the Hg profile in Lake Bosomtwe's sediments indicates

a strong decrease in atmospheric Hg deposition to the lake. This discrepancy may be related to the recently reported differences in trends in anthropogenic Hg emissions (which are likely increasing) and global atmospheric mercury concentrations (which were found to have declined by ~30% since 1995; Slemr et al., 2011). Alternatively, the decline in Hg flux to the Lake Bosomtwe sediments may reflect long-range atmospheric input from a region where Hg emissions are declining. Given the potential for atmospheric transport of European pollution to Africa (Kallos et al., 1998; Duncan and Bey, 2004; Kallos et al., 2007), the recent decline in Hg delivery to the sediments of Lake Bosomtwe may reflect trends in European emissions.

In both North America and Europe, it has been observed that lake sediments can be slow to reflect decreases in atmospheric deposition of mercury, particularly in lakes with high catchment area to lake area ratios (Engstrom et al., 1994; Kamman and Engstrom, 2002; Meili, 1995). After more than a century of elevated mercury deposition to these regions, there remains a large inventory of mercury in the terrestrial catchments of these lakes, which continues to be exported to the lake despite decreasing atmospheric inputs (Kamman and Engstrom, 2002). The rapidity of the decline in Hg flux to Lake Bosomtwe's sediments may reflect the extremely low terrestrial catchment to lake area ratio of the lake, which would allow Hg flux to Lake Bosomtwe's sediments to be highly sensitive to changes in atmospheric deposition of mercury while reducing the influence of terrestrially deposited Hg. Direct deposition of Hg to the lake surface dominates Hg inputs to Lake Bosomtwe, allowing Hg flux to the sediments to respond rapidly to and closely reflect atmospheric inputs.

Mercury flux ratios (ratios between estimated flux values for specific time periods) are particularly useful because they are directly comparable between lakes, since they are not influenced by differences in catchment size, catchment Hg export, or the magnitude of Hg flux. Based on our estimates, modern Hg flux to Lake Bosomtwe's sediments is 1.6-fold higher than 1870 flux, while peak flux was 3.0-fold higher than 1870 flux (Table 2, Fig. 3b).

Yang et al. (2010b) report that globally, most sediment records taken from lakes with no important local sources of Hg pollution show a 3-fold increase in Hg from pre-industrial to modern times. In Lake Bosomtwe, the ratio of modern flux relative to 1870 flux may be lower than what is reported as typical by Yang et al. (2010b) for several reasons, including: 1) calculation of flux ratio

Table 2
Reference (1870), peak and modern annual Hg flux to the sediments of the Central 1 coring site in Lake Bosomtwe. Flux ratios relative to 1870 flux are also shown for these time periods.

Time period	THg flux to Central 1 sediments ($\mu g\ m^{-2}\ y^{-2}$)	Flux ratio
Reference (1870)	13.5	1.0
Peak (1967)	40.5	3.0
Modern (2000)	20.9	1.6

based on a post-1850 reference date, 2) rapid response to recent declines in Hg deposition (due to a small catchment to lake area ratio), and/or 3) pre-industrial Hg contamination due to regional gold mining activities.

In particular, due to Ghana's long history of gold mining, estimated Hg flux to the sediments for 1870 (or even 1850) is unlikely to represent background conditions, and it is possible that our calculated flux ratios may underestimate the true human-mediated increase in Hg inputs to the lake. This issue has been highlighted by Cooke et al. (2009, 2010), whose work in the Peruvian Andes has shown pre-industrial mining-related increases in sediment Hg. Gold mining has been taking place in Ghana for over 1000 years (Hilson, 2002) with the arrival of Europeans in 1471 driving an increase in AGSM activities to meet extra-regional demand for gold (Donkor et al., 2006b). As such, future paleolimnological work may be helpful in determining the extent of pre-industrial Hg contamination in this region.

4. Conclusion

Lake Bosomtwe's sediments provide unique information about trends in historical and current mercury deposition to a region for which few data exist. As has been observed based on other cores from around the world, mercury inputs to Lake Bosomtwe increased throughout the industrial era. However, the recent strong decline in Hg flux to Lake Bosomtwe's sediments is not consistent with our understanding that Hg emissions are likely to be increasing on a local, regional, and global scale. Instead, this decline may reflect the dominant role played by long-range atmospheric Hg transport in determining Hg deposition in West Africa.

Acknowledgments

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