Historical Reconstruction of Mercury Pollution Across the Tibetan Plateau Using Lake Sediments

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The Tibetan Plateau is described as the “Roof of the World” averaging over 4000 m above sea level; it is remote, isolated, and presumed to be a pristine region. In order to study the history of atmospheric mercury (Hg) pollution and its spatial variation across the Plateau, lakes were chosen from three areas forming a north to south transect. Sediment cores were taken from three sites in each area and dated using the radionuclides $^{209}$Pb and $^{137}$Cs. Analysis of the cores yielded the first comprehensive Hg reconstructions for the Plateau, showing clear Hg pollution at all sites. The first indication of Hg pollution is much earlier than the onset of the industrial revolution in Europe, but the most significant pollution increase is from the 1970s, followed by a further marked increase from the 1990s. The mean post-2000 atmospheric pollution Hg accumulation rates for the sampling sites were estimated at between 5.1 and 7.9 µg m$^{-2}$ yr$^{-1}$. The increase in Hg pollution over the last few decades is synchronous with the recent economic development in Asia (especially China and India), and pollution Hg levels continue to increase. Furthermore, contemporary sediment Hg accumulation rate data are in broad agreement with Hg deposition values derived from global models that attribute pollution to sources mainly within southeast Asia. As most of the sites are exceptionally remote and situated above the atmospheric boundary layer, these results underline the need to understand the local Hg cycle in both regional and global context.

Introduction

Atmospheric transport and deposition of Hg has resulted in widespread contamination (1, 2), potentially impacting sensitive ecosystems and species (3). In the last century, severe damage to human health from the toxicity of Hg has also been reported (4). Concerns over Hg toxicity have resulted in a massive reduction in Hg emissions since the 1970s in Europe and North America (5, 6). However, Hg emissions have increased in China and India in recent years due to rapid economic development (6). Such recent increases have given rise to the concern that Hg and other pollutants may be contaminating remote ecosystems in Asia, including those on the Tibetan Plateau, through long-range transported air pollution. Many studies show long-term of Hg pollution in North America and Europe (1, 2, 7–12), however, equivalent data for Asia are more scarce and there is now a concern with respect to the increase in Hg input from this region to the global cycle. The Tibetan Plateau is one of the most remote and isolated regions in the world, and due to its high altitude and the size of its glaciers it is often referred to as the “Roof of the World” or the “Third Pole”. It is a unique landscape and the source of many major rivers in Asia. Hence, it is a source of water for half the world’s population and a fragile environmental resource that requires protection from contamination. As much of the Tibetan Plateau lies above the atmospheric boundary layer, assessing the extent of Hg contamination is also important in understanding both the regional and global Hg cycle. Due to the difficulty of sampling in the region, there have so far been few Hg studies. Loewen et al (13) and Wang et al (14) measured Hg in snowwips in order to estimate atmospheric Hg deposition fluxes. However Hg in snowwip can be affected by photoreduction resulting in a considerable proportion of the Hg being released to the atmosphere soon after the snow is deposited (15, 16). Since ultraviolet light enhances Hg release (15) and ultraviolet light is very strong on the Plateau, Hg deposition flux records determined from snowwip samples are both short-term and under-estimations.

Lake sediments, by contrast, are natural archives that provide an historical record of environmental change within a lake and its catchment as well as trends in atmospheric deposition to the lake surface. Analysis of samples taken from a sediment core therefore allows changes through time to be identified, whereas measurements of natural and artificial radionuclides allow a reliable sediment chronology to be constructed. In this way dates at which changes occur can be determined and rates of pollution deposition estimated. Sediment cores have been extensively used in this way over the past several decades to target the historical accumulation of Hg, primarily as a result of atmospheric deposition (7–10).

In order to study the historical record and spatial distribution of Hg across the Tibetan Plateau, sediment cores were collected from lakes in three areas along a north-to-south transect (Figure 1). Here, we assess the extent of Hg contamination across the Plateau and compare the results with simulations of Hg deposition from global Hg models.

Materials and Methods

Sediment Coring. Sediment cores were taken from the deep parts of selected lakes in summer 2006 for the northern and central areas of the Tibetan Plateau and in summer 2007 for the southern area (Figure 1) using a HTH gravity corer fitted with an 8.5 cm inner diameter polycarbonate tube. Cores from Nam Co and Peku Co were taken from the deepest part of shallower subbasins. The cores were sectioned in the field using a stainless steel slicer at 0.25 cm intervals from the surface to 5 cm, and then at 0.5 cm intervals from 5 cm to the base of the core. In the catchment of each lake, soil samples were taken down to 20 cm depth from the face of a freshly dug soil pit using stainless steel sampling implements. Soil sampling intervals were at 0.5 cm from the surfce to 5 cm, and then at 1 cm intervals from 5 to 20 cm.

The samples were stored at 4 °C until subsamples were measured for wet density, for water content by measuring...
mass loss after heating at 105 °C for 24 h, and loss-on-ignition (LOI), a proxy for organic matter content, by heating at 550 °C for 4 h. The samples were then freeze-dried, homogenized, radiometrically dated, and analyzed for Hg.

**Radiometric Dating.** Samples from the sediment cores were analyzed for 210Pb, 226Ra, 137Cs, and 241Am by direct gamma assay in the Bloomsbury Environmental Isotope Facility (BEIF) at University College London, using an ORTEC HPGe GWL series well-type coaxial low background intrinsic germanium detector. Sediment accumulation rates for the cores were calculated from 210Pb radiometric data (Supporting Information (SI) Figure S1). Soil cores were gamma counted in the same way as for the sediment cores in order to estimate atmospheric unsupported 210Pb deposition fluxes in the sampling areas.

**Mercury Analysis.** After the sediment cores were dated, samples were selected from each core in order to provide sufficient temporal resolution to show historical trends in Hg concentrations. Mercury samples were digested using 8 mL of conc. HNO₃ (Aristar) at 100 °C on a hot plate for 1 h in rigorously acid-leached 50-mL Teflon beakers. Digested solutions were analyzed for Hg using cold vapor-atomic absorption spectrometry (CV-AAS) following reduction with SnCl₂ (8, 9). External quality control was achieved by digesting and analyzing replicate standard reference material (stream sediment GBW07305) and analytical blanks. Average Hg recovery rates were 93 ± 8% for reference material GBW07305.

**Corrected Hg Accumulation Calculations.** Sediment focusing is a process that occurs in most lake basins (17) causing preferential accumulation of sediment in deep water. In order to calculate mean basin accumulation rates, a correction for sediment focusing can be made by dividing the unsupported 210Pb accumulation rate in a sediment core by the atmospheric 210Pb deposition flux for the region. In this study, unsupported 210Pb accumulation rates in the catchment soils were used as proxy measurements for the atmospheric 210Pb deposition flux ((18), SI Table S1). Corrected Hg accumulation rates (i.e., Hg accumulation rates corrected for sediment focusing) were calculated using these adjusted 210Pb data and used to make comparisons with simulations of Hg deposition from the global Hg model STOCHEM.

**The Global Hg Model.** Airborne Hg and wet Hg deposition fluxes over the Tibetan Plateau were calculated using the STOchastic CHEMistry (STOCHEM) model, details of which are given elsewhere (19). STOCHEM is a global Lagrangian chemistry-transport model to which, in this study, a Hg life cycle has been added based on Ryaboshapko et al. (20). Man-made Hg emissions amounting to 1276 tonnes per year have been taken from Wilson et al. (21). Natural Hg emissions from oceans and soils have been based on Bergan and Rodhe (22), with the latter source strength scaled up to produce model gaseous Hg concentrations over Europe and North America that agree with observations.

**Results**

**Sediment Accumulation in Tibetan Lakes.** Eight of the nine cores had good 210Pb and 137Cs profiles, suggesting the sediments had not been disturbed. However, the anomalous 210Pb profile of a core taken from a salt lake, Cuulong Co, indicated that the sediments in this core were not stratigraphically conformable and could not be used for dating. For this reason, results from the Cuulong Co core are not included in this paper. The equilibrium depths of total 210Pb with supported 210Pb (corresponding to an age of ca. 150 years) for the remaining eight cores are mostly deeper than 18 cm (Table 1), indicating that these cores have a sediment accumulation rate sufficient to provide a good temporal resolution for studying modern pollution trends.

In general, pre-1950 sediment accumulation rates were fairly stable in the cores, within a range of 0.01–0.05 g cm⁻² yr⁻¹ (SI Figure S1). After ca. 1950, some cores show a slight increase in accumulation rate, whereas other cores show a significant increase and even a peak in the 1970s and/or 1980s. The Cuo Na core is unusual. There is a defined accumulation rate peak in ca. 1965 and exceptionally high values that reach 0.8 g cm⁻² yr⁻¹ in 2004. This is probably caused by railway construction by the lake as peak ac-
cumulation is followed by a sharp decline after the date at which railway construction was completed.

Since sediment accumulation rates in all the cores were stable before 1950, we have used the basal sediment accumulation rates from each core to extrapolate dates back in time to give approximate ages for the older pre-210Pb sediments.

**Mercury Concentrations.** The Hg concentration profiles (Figure 2) show a general increasing trend from the deeper parts of the cores upward to the surface. An increase in Hg concentrations before the 1850s, the onset of the industrial period in Europe, is clear in some cores. For example, in the Keluke and Peku Co cores the increase occurs in the 17th century and in the Qinghai and Nam Co cores it occurs in the 18th century. Increases in Hg concentration occur through to the sediment surface in all cores except that from Keluke which shows a decline in Hg concentration in recent decades (Figure 2). This may be due to dilution caused by a rapid increase in sediment accumulation rates due to deeply incised erosion of the catchment over this period (SI Figure S1).

**Mercury Accumulation Rates.** Mercury accumulation rates calculated by considering variation in sedimentation rates, as described in Perry et al. (23) reveal a slow increase for some cores from lowest sediment levels, whereas others show relatively constant rates before 1950 (Figure 3). Since the 1970s, there has been a significant increase in Hg accumulation rates at all sites and after 1990, this increase becomes more rapid, especially in southern and central areas of the Plateau. Rates in the Cuo Na core over the last ca. 40 years are much higher than those in the other cores. This is probably due to local road and railway construction within the catchment and it is assumed that this disturbance caused Hg-containing soils to be washed into the lake.

Geochemical element concentrations and organic matter content are fairly constant in the lower portion of the cores, suggesting relatively stable natural sources over these early periods. Some cores show an increase in Hg concentrations prior to the 1860s, suggesting possible early pollution. However, the mean Hg concentrations at the base of the sediment cores are closest to, or equal to, background levels. Hence, pollution Hg was estimated using the mean Hg concentrations at the base of the sediment cores as a background level. Correcting these data for sediment focusing (see Methods section) to estimate pollution Hg accumulation rates (23) on a lake-basin scale shows that Hg contamination in these sites was very low prior to 1950, but since then, Hg pollution has rapidly increased (Figure 4).

**Modeled Fluxes.** Output from the global Hg model STOCHEM for the three 5° × 5° grid squares covering the location of the lake sites in the Tibetan Plateau are shown.
The wet deposition flux from pollution sources is estimated to be 5.1, 3.7, and 3.3 \( \mu \text{g m}^{-2} \text{yr}^{-1} \) for the southern, central, and northern grid squares, respectively. This may be compared to the mean pollution accumulation rate from the surface sediments that vary from 5 to 70 \( \mu \text{g m}^{-2} \text{yr}^{-1} \) (Table 2). The model also indicates that although Asia is the main pollution source, the Plateau also receives long-range transported Hg from all other continents (SI Table S2).

**Discussion**

**Mercury Inputs and Corrected Accumulation Rates.** Mercury emissions to the atmosphere from anthropogenic sources have a long history (5, 24), increasing substantially since the onset of the industrial period in Europe and resulting in an increase in atmospheric Hg deposition. For very remote lakes nearly all the inputs of Hg are likely to be derived from direct atmospheric deposition to the lake surface although some inputs can be expected to be routed via the catchment primarily from the upper soil horizon during periods of high runoff (cf., ref 25). The quantities involved depend on the catchment area (11) and nature of the soils (26) as well as runoff characteristics. Mercury concentrations in most of our Tibetan catchment soil samples are low, in the range of 7–16 ng g\(^{-1}\), and the contribution of Hg from the erosion...
of these soils can be assumed to be small (cf., ref 26). Therefore, atmospheric Hg deposition should be the main Hg source for these lakes, especially over recent times. Furthermore, the studied lakes can be expected to retain Hg inputs because nearly all the lakes occupy closed basins and have no distinguishable outflows. As Hg loss through water–air interface exchange is relatively low compared with fluxes to the sediments (27), it can be assumed that, aside from volatilization, Hg losses are minimal and that input into the lakes is retained and deposited to the sediments.

Mercury deposited within the lake basin can be redistributed by sediment focusing and we have used the 210Pb record to correct for this. The post-2000 corrected Hg accumulation rates for five of the sites (i.e., all except for Keluke, Cuo Na and Peku Co) are between 10.4 and 16.1 µg m⁻² yr⁻¹ with a mean of 12.6 µg m⁻² yr⁻¹ (relative standard deviation (RSD) = 2.3 µg m⁻² yr⁻¹). The similarity of these values supports the view that they mainly represent atmospheric deposition fluxes. Differences between these sites may be related to individual catchment influences and/or to uncertainties in the correction. The higher values for Keluke may be related to its use as a fishery and values for Cuo Na have been affected by railway construction as described above. Peku Co lies at the base of Mt Xixiabangma and receives glacial meltwater through the subbasin where the core was taken to the open lake and the meltwater may contain Hg previously deposited and stored in glacier ice. Some of the other sites in the region, such as Nam Co, also receive glacial meltwater, but at Nam Co the core was taken in a bay in the southeast of the lake removed from inflows influenced by meltwater inputs.

For the five lakes not affected by direct human activity or glacial meltwater we assessed the influence of their catchments by relating the post-2000 corrected Hg accumulation rates (µg m⁻² yr⁻¹) to their catchment-lake area ratios (x), with the following result (SI Figure S3):

\[ y = 11.2 + 0.062x (R^2 = 0.36) \]

The intercept of the regression in this relationship indicates that the accumulation rate value for a lake with no terrestrial catchment is 11.2 µg m⁻² yr⁻¹. This compares with a value of 12.6 µg m⁻² yr⁻¹ for the mean accumulation rate of the five sites and suggests that approximately 89% of the Hg in the sediments is derived from direct atmospheric bulk deposition (including dust, see below). The catchment impact factor (0.062) is a much lower figure than that observed by Swain et al. (11) in North America. There are two main reasons that probably explain the relatively low contribution of catchment sources. First, the combination of low rainfall on the Tibetan Plateau (250–500 mm yr⁻¹ (28)), and sandy catchment soils means that runoff by rainfall is not easily formed and hence Hg transport by runoff is very low. Further, Hg is not easily transported through these soils to the lakes (29). Second, because of the high altitude, most wet deposition on the Plateau is in the form of snow. It has been observed that most Hg²⁺ deposited in fresh snow may be rapidly photoreduced to gaseous elemental mercury and then released back to the atmosphere (15, 16). This release may be enhanced by the strong ultraviolet light (15) on the Plateau.

The post-2000 corrected pollution Hg accumulation rates for the lakes were calculated using the mean pollution Hg values in the sediments formed since 2000 as shown in Table 2. If the values for Keluke, Cuo Na, and Peku Co are excluded for the reasons outlined above, the pollution Hg accumulation rates for the undisturbed lakes tend to have similar values, between 5.1 and 7.9 µg m⁻² yr⁻¹ with a mean of 6.3 µg m⁻² yr⁻¹ (RSD = 1.1 µg m⁻² yr⁻¹). This further reinforces the view that these accumulation rate values represent atmospheric pollution Hg fluxes and may be reasonably expected to be similar across the Plateau. We can therefore compare these pollution values with other estimates of Hg deposition flux from snowpits and from modeling.

Snowpits have recently been used to measure atmospheric Hg deposition on the Tibetan Plateau. Published values differ markedly and range from either 0.74 – 2.97 µg m⁻² yr⁻¹ (13) to 6 – 18 µg m⁻² yr⁻¹ (14). However, as Hg in the snow can be quickly released back to the atmosphere (15, 16), snowpit values are likely to be variable and in general underestimate the deposition flux. Estimated background atmospheric Hg deposition using sediments from midcontinental North America (11) gives a value of about 3.8 µg m⁻² yr⁻¹, slightly lower than our Tibetan Plateau lake sediment values (4.9 µg m⁻² yr⁻¹). However, Hg deposition on the Tibetan Plateau can be strongly affected by dust (13). As the quantity of dust may be surprisingly high and duststorms are able to reach altitudes of ca. 10 km above sea level on the Plateau (30), dust may play an important role in natural Hg deposition to these lakes. Wet deposition flux values for Hg using the global mercury model STOCHEM (SI Figure S2) are consistently lower than the calculated accumulation rates from the sediment cores. This may indicate the presence of a possible local contribution from the resuspension of windblown dust which is not included in the modeling.

**Enrichment Factors in Hg Accumulation.** In the last ca. 150 years, Hg deposition has increased globally about 3-fold (31), and this has been reflected in records from lake sediments, peat deposits and glacial ice at remote sites in both the northern and southern hemispheres (1). The extent of the recent contamination can be assessed by comparing accumulation rates at the present day with those in the 1850s. Factors for the increase in Hg accumulation rate are 4.7 and 5.0 in the southern area and 8.2, 10.9, and 12.7 in the central area of the Plateau (the 12.7 value for Cuo Na may have been affected by the increased sedimentation rate caused by the railway construction in the catchment). However, these values

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**TABLE 2. Mercury Accumulation Rate Data, Focussing Factors and Enrichment Factors for Hg Accumulation in the Cores (M/P = Modern to Preindustrial Hg Ratio)**

<table>
<thead>
<tr>
<th>lake</th>
<th>post 2000 Hg accumulation rate (µg m⁻² yr⁻¹)</th>
<th>1860 Hg accumulation rate (µg m⁻² yr⁻¹)</th>
<th>M/P</th>
<th>post 2000 pollution Hg accumulation rate (µg m⁻² yr⁻¹)</th>
<th>focusing factor</th>
<th>corrected Hg accumulation rate (µg m⁻² yr⁻¹)</th>
<th>mean corrected pollution Hg accumulation rate (µg m⁻² yr⁻¹)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Qinghai</td>
<td>15</td>
<td>5.3</td>
<td>2.8</td>
<td>9.6</td>
<td>1.36</td>
<td>11.0</td>
<td>7.0</td>
</tr>
<tr>
<td>Keluke</td>
<td>28</td>
<td>1.9</td>
<td>14.7</td>
<td>15.4</td>
<td>0.72</td>
<td>38.8</td>
<td>21.3</td>
</tr>
<tr>
<td>Gaihai</td>
<td>17</td>
<td>5.4</td>
<td>3.2</td>
<td>6.3</td>
<td>1.24</td>
<td>15.3</td>
<td>5.1</td>
</tr>
<tr>
<td>Cuo Na</td>
<td>434</td>
<td>34.0</td>
<td>12.7</td>
<td>125.0</td>
<td>5.97</td>
<td>72.7</td>
<td>20.9</td>
</tr>
<tr>
<td>Cuo E</td>
<td>73</td>
<td>8.9</td>
<td>8.2</td>
<td>37.8</td>
<td>6.26</td>
<td>11.7</td>
<td>6.0</td>
</tr>
<tr>
<td>Nam Co</td>
<td>16</td>
<td>1.5</td>
<td>10.9</td>
<td>12.6</td>
<td>1.58</td>
<td>10.4</td>
<td>7.9</td>
</tr>
<tr>
<td>Peku Co</td>
<td>23</td>
<td>4.6</td>
<td>5.0</td>
<td>12.0</td>
<td>0.17</td>
<td>133.9</td>
<td>69.9</td>
</tr>
<tr>
<td>Kemen Co</td>
<td>30</td>
<td>6.4</td>
<td>4.7</td>
<td>10.2</td>
<td>1.87</td>
<td>16.1</td>
<td>5.5</td>
</tr>
</tbody>
</table>
are considerably higher than the 3-fold factor observed in other remote sites (cf 1, 7, 8, 32).

Mercury exists in different forms in the atmosphere. Although gaseous elemental mercury (GEM) is the dominant form, divalent reactive gaseous mercury (RGM) is water-soluble, and can be rapidly bound to particle surfaces or scavenged by rain and cloud droplets before dry and wet deposition (33, 34). For GEM to be converted to RGM, atmospheric oxidants are required. The lakes on the Plateau, especially those in the central and southern areas, are at altitudes over 4500 m above sea level and frequently exposed to free tropospheric air, where oxidant concentrations are higher, especially during winter. Further, an increase in atmospheric oxidants in tropospheric air since preindustrial times has been reported (35). The RGM formation, and subsequent Hg deposition may be expected to increase with altitude and there is good evidence that this is so (34, 36). This may explain the higher Hg enrichment factors in these regions of the Plateau. The lower enrichment factors in the southern area may due to the influence of winds derived from low altitude with lower oxidant levels while tropospheric air dominates the central part of the Plateau.

By contrast the Hg enrichment factors in the Qinghai and Ghai cores taken from the northern area are approximately 3-fold, suggesting that Hg deposition in this area may still be controlled by boundary layer air, even though the lakes are around 3000 m above sea level. The core from the third lake in the northern area, Keluke, has an anomalously high accumulation rate increase of 14.7, and this is possibly due to recent local contamination from fisheries in the lake.

Mercury Sources. Although the main increase in Hg concentration in Tibetan Plateau lake sediments postdates 1850, there is also evidence for earlier contamination (Figure 2, SI Figure S4). It is likely that early contamination in such remote basins is the result of pollution from local or regional sources (e.g., refs 10 and 12) rather than global ones. In Tibet, Hg has been used in different ways for centuries. For example, vermillion has been used in Tibetan painting and decoration (37), and Hg has also been used in traditional Tibetan medicines (38). There is evidence that both vermillion and Hg were exported from India to Tibet during the 18th and 19th centuries (39). In addition, traditional cooking with wood, dung and crop residues (40) and butter burning for religious purposes might also have been sources of Hg to the atmosphere.

Since the onset of the industrial revolution in Western Europe, human activities have significantly increased atmospheric Hg emissions, deposition and global Hg contamination (5). The atmospheric lifetime of Hg may vary, but its average residence time is sufficiently long for Hg emitted by developed industrial countries to have reached the Tibetan Plateau (13, 41). Emissions from industrial countries increased rapidly from 1850 until the 1970s, after which there has been a rapid decline as a result of emissions reductions policies implemented due to wide concerns about Hg toxicity (5). The increase in Hg concentration in Tibetan Plateau sediments before 1970 but after 1850 is likely therefore to have been derived from global industrial sources. However, the corrected pollution Hg accumulation rate values suggest that, in general, the atmospheric fluxes to the sites were relatively small during that time and the increase in flux rate before the 1950s is slow when compared to recent years.

The beginning of the rapid increase in Hg accumulation rates at most sites occurs in the 1970s, followed by a further increase from the 1990s up to the present day such that pollution Hg fluxes in recent years are much higher than the levels before 1950. Mercury emissions from developed countries have been dramatically reduced since the 1970s (e.g., ref 5), resulting in reduced atmospheric Hg deposition (42). However, Hg emissions in Asia have increased due to the increase in coal combustion in the region (4, 5, 43) related to rapid economic development since the 1970s. It is estimated that Hg contribution from Asian countries to worldwide total Hg emissions in 2000 was 54%, an increase from 30% in 1990 (6, 44).

China was the first and India the third highest Hg emitting countries in the world in 2000 (6). Streets et al. (45) have calculated that for China, 32% of emitted Hg is released as Hg(0), and 12% as particulate Hg. These emissions tend to be deposited locally and regionally. However, most emitted Hg (56%) in China is as GEM, and GEM has the potential for long-range transport. The sites in this study are located at high altitude, and most are likely to be in the free troposphere. At these altitudes deposited Hg is likely to be derived from long-range sources, probably mostly from industrialized areas of Eastern China and Northern India (46). On the Tibetan Plateau precipitation is strongly controlled by the Asian monsoon regime (47) including the southwest monsoon from India and the east Asian monsoon from eastern China and both have the potential to transport Hg to the Plateau. The dominance of Asian sources is indicated by the global Hg model STOCHEM (SI Table S2), although the model indicates that Hg is also derived from more distant sources, illustrating the fully global nature of Hg pollution at the present day. The increasing trend in pollution Hg in the 1970s and 1980s in Tibetan sediments is very similar to that in UK sediments in the mid-19th century which represented the beginning of industrial Hg pollution in Western Europe (10). Hence, the rapid increase in pollution Hg accumulation rates in the 1970s and 1980s in Tibetan sediments may represent an impact from the beginning of a “new industrial revolution” in Asia.

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Supporting Information Available

Two tables and four figures. This material is available free of charge via the Internet at http://pubs.acs.org.

Literature Cited


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