

Relationship between mercury and organic carbon in sediment cores from Lakes Qinghai and Chenghai, China

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Abstract

Purpose Factors such as organic matter can significantly influence the distribution of mercury (Hg) in aquatic environments. Recent studies in Arctic and sub-Arctic lakes in Canada have investigated whether scavenging of Hg by phytoplankton significantly affects distributions of Hg in sediments. This study examined the relationships between Hg and organic components in two contrasting lakes (Lakes Qinghai and Chenghai) in low and middle latitudes of China.

Materials and methods Sediment cores from the less-polluted, oligotrophic Lake Qinghai (QH) and from the polluted, eutrophic Lake Chenghai (CH) were collected by a gravity corer. The cores were sectioned and transported on ice to the laboratory where they were stored at -20°C . Subsamples were dried in a vacuum freeze dryer and grounded with a mortar and pestle prior to analyses. Total concentrations of Hg were quantified using cold vapor atomic absorption spectrometry. Total organic carbon (TOC) was quantified using an elemental analyzer after removal of carbonate. The Rock-Eval 6 pyrolysis technique (Vinci Technologies, Rueil-Malmaison, France) was used to

deconvolute TOC in sediments into S1, S2, and RC components; S2 was further separated into S2a and S2b.

Results and discussion Different relationships between Hg and TOC were found in the two lakes, which suggest that different types of organic compounds might play completely different roles in the distribution of Hg in lakes. S1 (the soluble organic matter (SOM)) was found to significantly control distributions of Hg in sediments of both lakes, while S2 and S2a were not. Combining the synchronous fluctuations of Hg and the oxygen index in the QH sediment core and in recent sections of CH suggested that allochthonous SOM derived from the terrestrial environment had an important influence on the distribution of Hg in both lakes and a large portion of Hg that originated from the lake catchment.

Conclusions This study provides further evidence that organic matter is one of the most important factors that influences distributions of Hg in lake sediments and that SOM was the primary form of carbon associated with sedimentation of Hg. The results also suggest that Hg in lake sediments might not accurately represent its pollution history as it could also be influenced by land use, such as agriculture or other human activities in the catchment.

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1 Introduction

Mercury (Hg) is a naturally occurring element that comes from a range of natural sources (Joensuu 1971; Poissant 1999; Seigneur et al. 2003; Parsons et al. 2007). Mercury, being a soft electron donor, is often associated with sulfur while it cycles naturally in the lithosphere or through mobilization from materials stored deeper in the crust of the Earth, such as the burning of fossil fuels, especially coal, and the smelting of metals. The use of Hg in various human

activities has resulted in mobilization of greater concentrations of Hg into the lithosphere, where it can interact with humans and wildlife. Mercury can cause toxic effects at ecologically relevant concentrations and can be transported long distances in the atmosphere. Mercury accumulates in the sediments of lakes, which can, in turn, serve as a historical record of Hg in the environment. However, factors such as organic matter might significantly influence distributions of Hg in aquatic environments (Lindberg and Harriss 1974; Kainz et al. 2003; Mirlean et al. 2003; Kainz and Lucotte 2006; Sanei and Goodarzi 2006; Outridge et al. 2007; Parsons et al. 2007; Carrie et al. 2009). Significant associations between total organic carbon (TOC) and the distribution of Hg have been observed in sediments (Kainz et al. 2003; Mirlean et al. 2003; Kainz and Lucotte 2006). The results of recent studies as to whether scavenging of Hg by phytoplankton is a significant factor in the distribution of Hg in sediments (Sanei and Goodarzi 2006; Outridge et al. 2007, 2011; Stern et al. 2009; Deison et al. 2012; Kirk et al. 2011, 2012; Cooke et al. 2012). The Rock-Eval and petrographical results (Sanei and Goodarzi 2006) for eutrophic lakes of central Alberta, Canada found that correlations between Hg and TOC were due primarily to the labile portion of organic C (S1), which consists of more easily degradable lipids and pigments derived from algae. Significant correlations between concentrations of Hg in sediments and algal-derived organic matter were observed in Canadian Arctic and sub-Arctic lakes (Outridge et al. 2007; Stern et al. 2009). Based on the relationships between Hg and S2 before 1900, the results of predicting Hg fluxes in arctic lakes after 1900 indicated that significant portions of concentrations of Hg in sediments could be explained by scavenging by aquatic primary productivity during the past decades and centuries, but not by human activities, especially during the latter half of the twentieth century. These correlations led to the hypothesis that recent records of Hg in sediments of higher latitude lakes could have been confounded by scavenging of Hg from the water column by algae so that Hg concentrations or fluxes observed in sediments might not accurately represent a historical deposition of Hg (Stern et al. 2009). However, a recent comparative study in 14 Canadian Arctic and sub-Arctic lakes (Kirk et al. 2011) suggested that scavenging by algae was not an important process governing Hg fluxes to sediments because some Arctic lakes were simultaneously experiencing greater algal abundance and lesser deposition of Hg. Spatial and temporal associations between Hg and organic C was investigated in 14 Arctic lakes in Canada (Deison et al. 2012) and suggested that organic matter derived from algae might be sources of Hg to sediments. However, other factors such as rate of sedimentation of inorganic materials and rate of erosion in the catchment are likely to mediate this effect (Deison et al. 2012). Whether scavenging by algae is

important in other places, such as lakes at lower latitudes, or other organic components might also be associated with deposition of Hg in sediments needs further investigation.

In this study, the relationships between concentrations of Hg and TOC in sediments were investigated in two lakes in China: one remote, less polluted, oligotrophic lake at higher latitude (Lake Qinghai (QH)) and a more contaminated, eutrophic lake at lower latitude (Lake Chenghai (CH)). The Rock-Eval pyrolysis technique was used to separate sediment organic C into S1, S2, and RC fractions, in which S2 was further separated into S2a, which comprised thermally less stable macromolecular organic matter, and S2b, which comprised larger molecular weight kerogens (Xu et al. 2011b; Wu et al. 2012). In addition, the relationships between Hg and the organic matter (OM) derived from algae and the soluble organic matter (SOM) were investigated.

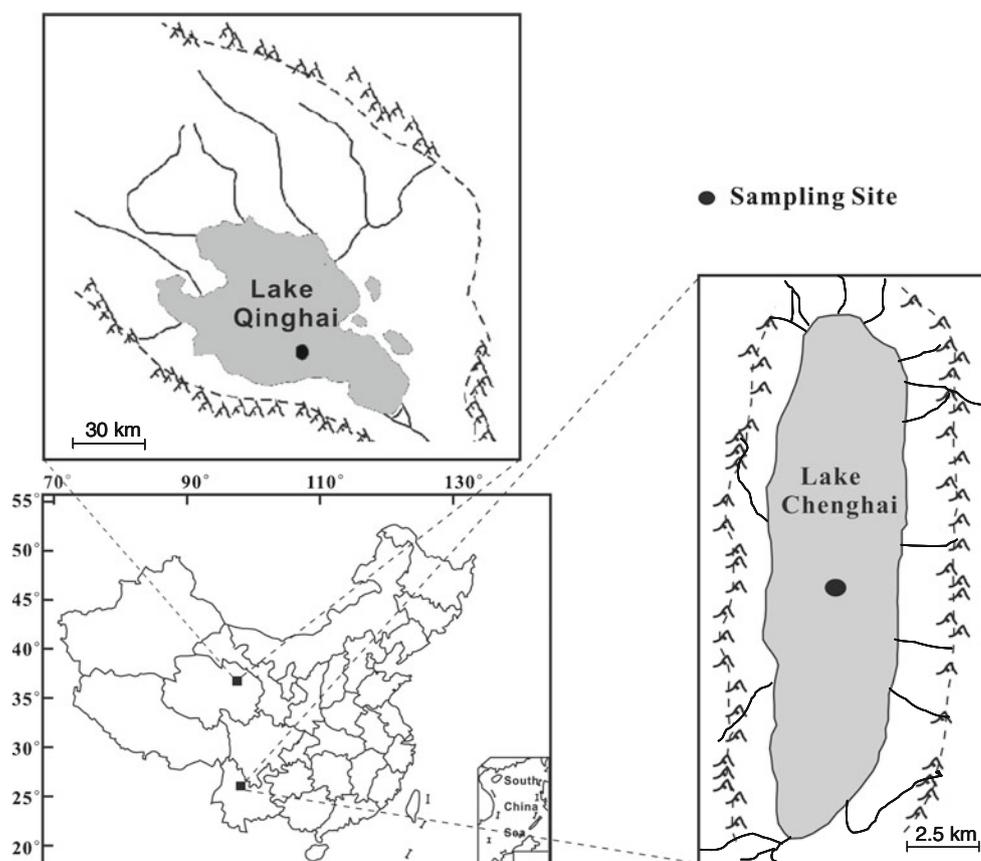
2 Samples and methods

Two sediment cores (QH and CH) were collected in 2006, one from the center and deepest area of each lake (Fig. 1). Lake Qinghai (36°32'N–37°15'N, 99°36'E–100°16'E) in the northeast of the Qinghai Plateau is the largest inland lake in northwestern China with an area of 4,583 km². It is a salty lake surrounded by mountains. The catchment area is 29,661 km². Its main water supplies are from precipitation and meltwater from snow and ice. There are no large urban centers in close proximity to the lake. The trophic status of the lake is oligotrophic (Yang et al. 2008). Lake Chenghai (26°27'N–26°37'N, 100°38'E–100°41'E) is located in Yongshen town in the Yunnan-Guizhou plateau of southwestern China, 50 km southeast from Lijiang City. It is a nutrient-enriched lake (Wan et al. 2005) with an area of 77.2 km², and the catchment area is 228.9 km².

Sediment cores were collected using a Teflon gravity corer. Once the cores were collected, sediment was extracted (ca. 1 cm) and immediately sliced using a Teflon circle, and wrapped in aluminum foil that had been baked (450 °C). Sectioned cores were transported on ice to the laboratory, where they were stored at –20 °C until further treatment. Subsamples were lyophilized and ground with a mortar and pestle before analyses.

Total concentrations of Hg in sediments were quantified at Trent University, Ontario, Canada by cold vapor atomic absorption spectrometry. Quality assurance was performed on certified reference materials (NIST 1633b and 2709), and results were in accordance with the standards. TOC in sediments was quantified using an elemental analyzer after the removal of carbonate. Reproducibility was within 3 % for TOC. The organic C in bulk sediments was operationally characterized by the Rock-Eval 6 analysis (Vinci Technologies, Rueil-Malmaison, France) at the State Key

Fig. 1 Locations of sampling sites in Lakes Qinghai and Chenghai, China. *Solid line* in the maps of the lakes represents inflowing and outflowing rivers; the *dashed line* represents the catchment boundary



Laboratory of Organic Geochemistry of Guangzhou Institute of Geochemistry, Chinese Academy of Sciences. This procedure allowed the quantification of three types of sedimentary organic C based on the thermal evolution of hydrocarbons, which are separated into S1, S2, and RC carbon groups (Lafargue et al. 1998; Sanei et al. 2005). The samples were first pyrolyzed in an inert atmosphere (He) from 100 to 650 °C at a rate of 25 °C min⁻¹. S1 was the hydrocarbon released under 300 °C, and S2 was the hydrocarbon released between 300 and 650 °C. The amounts of CO₂ (S3 in mg CO₂ g⁻¹) and CO (S3CO in mg CO g⁻¹) produced during pyrolysis were also measured. Samples were then automatically transferred to an oxidation oven and were heated from 400 to 850 °C to incinerate all the residual C (RC). A standard reference material (IFP 160000, VINCI Parc d) was used in the Rock-Eval measurements. In sediments, S1 originates from the pyrolysis of mainly lower molecular weight amorphous C, including free hydrocarbons, readily volatilized plant geolipids, and humic acid-derived hydrocarbons (Sanei et al. 2005; Outridge et al. 2007). S2, which is strongly associated with aquatic plants, may be a good indicator of aquatic primary productivity (Outridge et al. 2007). RC (or dead C) includes the final thermostable fraction of organic C from algae, terrestrial cellulose, and black C (Outridge et al. 2007; Carrie et al. 2009), which are rich in condensed aromatic C. The

hydrogen index (HI) is calculated as (S2/TOC)×100, while the oxygen index (OI) is calculated by normalizing the quantity of pyrolyzable CO₂ to TOC ((S3/TOC)×100). Statistical analyses were conducted using SigmaPlot 11.0 and SPSS 13.0, in which SPSS 13 was used to test the correlations (Pearson correlation).

3 Results and discussion

3.1 Concentrations of Hg and TOC

Profiles of concentrations of Hg and TOC in sediments of QH and CH are shown in Fig. 2. In core QH, the mean concentration of Hg was 43±14 ng Hg g⁻¹ (all measurements are dry mass) with the lowest concentration (23 ng Hg g⁻¹) at the bottom of the core and the greatest concentration (79 ng Hg g⁻¹) at the sediment surface. The profile of concentrations of Hg in core QH had an overall increasing trend from the bottom to the surface, especially in the top 5 cm, in which concentrations of Hg increased from approximately 50 to 80 ng g⁻¹. In core CH, the mean concentration of Hg was 96±61 ng Hg g⁻¹, with the lowest concentration (40 ng Hg g⁻¹) occurring at a depth of 74 cm, near the bottom of the core, and the greatest concentration (401 ng Hg g⁻¹) occurring at 10 cm depth. The concentration of Hg in the sediment of CH was approximately 2.2-fold

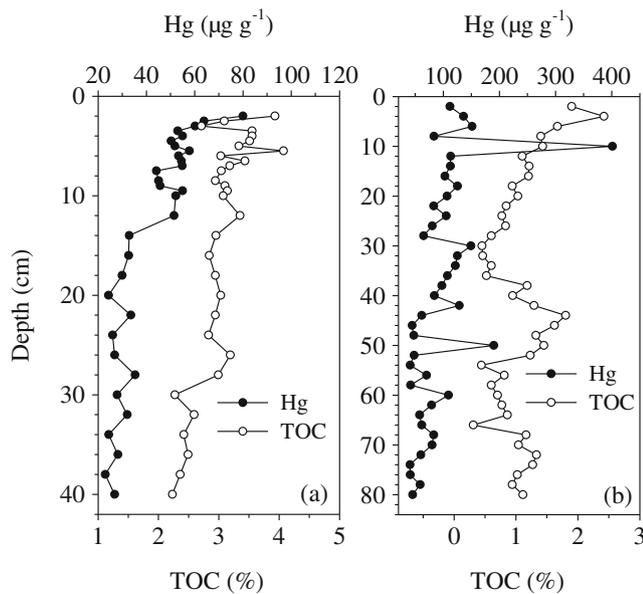


Fig. 2 Profiles of concentrations of mercury (*Hg*) and total organic carbon (*TOC*) in the sediment cores collected from Lakes Qinghai (a) and Chenghai (b)

greater than that in CH. The sediment accumulation rates were relatively constant in core QH (with a mean value of $0.017 \text{ g cm}^{-2} \text{ year}^{-1}$) (Xu et al. 2011a) and CH (with the mean value of $0.43 \text{ g cm}^{-2} \text{ year}^{-1}$) (Wan et al. 2005). Thus, fluxes of Hg to sediments in CH were greater than those in QH. These differences are related to human activities in the two areas. Southwestern China has a number of deposits of various metals, which include the largest deposit of Hg in China (Wanshan mercury deposit) and the city of Panchihua which is famous as a center for iron exploitation and smelting. Thus, Hg in sediments of CH is influenced by local industrialization through atmospheric deposition to the catchment of CH, while QH is located in northwestern China with relatively little influence from local industrialization or urbanization. Therefore, the flux of Hg into the sediments of CH was greater than that in QH.

The TOC content of core QH ranged from 2.23 to 4.07 %, with an overall increasing trend from the bottom to the top. Concentrations of Hg and TOC in the sediment core of QH were significantly correlated ($r=0.734$; $p<0.01$) (Table 1, Fig. 2). This result is consistent with similar associations observed in other studies (Lindberg and Harriss 1974; Regnell et al. 1997; Johannesson et al. 2003; Kainz et al. 2003; Mirlean et al. 2003; Kainz and Lucotte 2006). The significant correlation between TOC and Hg in core QH provides further evidence that organic matter plays a role in transporting and incorporating Hg to sediments. In core CH, the TOC content ranged from 0.31 to 2.24 % and the profile could be divided into three sections based on the TOC content depths of 80–54 and 52–30 cm, with the TOC having overall decreasing trends; from a 30-cm depth to the surface, it had an overall increasing trend. In core CH, the TOC profile was quite different from the profile of Hg

concentrations, and the correlation coefficient was 0.212 ($p>0.05$). The difference between relationships between TOC and Hg in the two sediment cores could be due to different types of organic C in the sediments of the two lakes (Ravichandran 2004; Sanei and Goodarzi 2006). In order to examine relationships between organic compounds and Hg, the TOC was divided into several types of organic C with different characteristics using the Rock-Eval technique.

3.2 Rock-Eval pyrolysis of sediment

Typical pyrolysis curves of sediments in cores QH and CH (Fig. 3) exhibited two peaks in the S2 group, which is similar to the pattern in sediments from Lakes Bosten, Hongfeng, and Shuangta in China (Xu et al. 2011b; Wu et al. 2012). Based on previous works (Xu et al. 2011b; Wu et al. 2012), organic C in QH and CH was separated into fractions S1, S2a, S2b, and RC, of which S2a should be a better indicator of primary productivity within a lake (Xu et al. 2011b; Wu et al. 2012). The S2a fraction, which is also referred to as R400, is mainly derived from algae (Carrie et al. 2012). Profiles of organic C (S1, S2a, S2b, and RC) in QH and CH are shown in (Fig. 4). In QH, concentrations of S1 ranged from 0.76 to $2.02 \text{ mg HC g}^{-1}$ with concentrations increasing from the bottom to the top of the core where the concentration was approximately 2.5 times greater than that at the bottom. Concentrations of S2a ranged from 1.00 to $2.32 \text{ mg HC g}^{-1}$ with a relatively stable profile in the sediment core. Concentrations of S2b and RC ranged from 2.37 to $5.51 \text{ mg HC g}^{-1}$ and from 1.60 to 3.13 %, respectively. Concentrations of both S2b and RC increased from the bottom to the top of the core. In CH, concentrations of S1, S2a, and S2b were 0.12 to 1.39, 0.26 to 2.59, and 0.28 to $2.69 \text{ mg HC g}^{-1}$, respectively. Profiles of concentrations were similar, and concentrations were greater near the surface. Concentrations of RC ranged from 0.20 to $1.85 \text{ mg HC g}^{-1}$ with a similar profile as that of TOC (see Fig. 4). Although concentrations of organic C in QH were slightly greater than those in CH, when differences in sediment accumulation rates were accounted for, fluxes of organic components in QH were less than those in CH. This observation is consistent with the trophic conditions of the two lakes: oligotrophic in QH and eutrophic in CH.

3.3 Relationships between Hg and organic carbon

Associations between Hg and organic C were observed throughout the history represented in the sediment cores (see Table 1). Mercury was significantly correlated with S2 in sediments of QH ($r=0.681$, $p<0.01$, $n=31$), but not in CH. This result suggests that S2 might play different roles in controlling the Hg distribution in the two lakes. However, when the relationships between Hg and mainly algal-derived S2a, which could be a better indicator of lake primary productivity (Xu et al. 2011b; Carrie et al. 2012;

Table 1 Correlation coefficients between mercury (Hg) and various organic components (S1, S2, S2a, S2b, RC, and TOC) in sediments of Lake Qinghai ($n=31$) and Chenghai ($n=40$)

| Lake | S1 | S2 | S2a | S2b | RC | TOC |
|----------|---------|---------|-------|---------|---------|---------|
| Qinghai | 0.848** | 0.681** | 0.343 | 0.702** | 0.586** | 0.734** |
| Chenghai | 0.358* | 0.238 | 0.267 | 0.206 | 0.158 | 0.201 |

See text for definition of organic components

*Significant at $p < 0.05$

**Significant at $p < 0.01$

Wu et al. 2012), were examined, no significant correlations were observed in both lakes. This result indicates that productivity of algae might not be the main factor which controls the distribution of Hg in sediments of CH and QH. Fraction S2, which represents productivity of algae in arctic lakes, had the greatest influence on the distribution of Hg in sediments of Arctic and sub-Arctic lakes in Canada (Outridge et al. 2007; Stern et al. 2009). In QH, components S1, S2b, and RC were all significantly correlated with concentrations of Hg (S1 $r=0.848$, $p < 0.01$; S2b $r=0.702$, $p < 0.01$; RC $r=0.586$, $p < 0.01$, with $n=31$ for all three). In CH, only S1 was significantly correlated with concentrations of Hg ($r=0.358$, $p < 0.05$, $n=40$). The common characteristic between the two lakes was the statistically significant correlation between the proportion of S1 and concentrations of Hg. Although the correlation coefficients between S1, S2, S2b, RC, and Hg were all statistically significant in QH, S1 exhibited the strongest correlation. In CH, even though atmospheric deposition of Hg was influenced more by human activities, there was still a significant correlation between Hg and S1 (see Table 1). Since soluble organic C (S1) might be influenced by degradation, it must be considered in the near-surface sediment. The pre-

1900 relationships between Hg and S1 were examined in QH (pre-1900 was below 5-cm depth in core QH, see Xu et al. 2011a), and the correlation coefficient was 0.859 ($p < 0.001$). In CH, the ratio of S1 to RC was relatively stable except for the top 4-cm sediment (Fig. S1, Electronic Supplementary Material), which indicated that S1 was relatively stable below the 4-cm depth of sediment. Mercury was also significantly correlated with S1 below the 4-cm depth in CH ($r=0.473$, $p=0.002$, $n=40$). These results demonstrated that, among the organic fractions, S1 was probably the most significant factor controlling the distribution concentrations of Hg in sediments of both CH and QH. Completely different relationships between S1, S2a, and Hg (S1 was significantly correlated with Hg, but S2a was not) suggested that algal scavenging was not the dominant factor which influenced the distribution of Hg in lakes at lower latitudes in China. Even though QH is located in a remote area with few industrial influences, which is similar to Arctic and sub-Arctic lakes (Outridge et al. 2007; Stern et al. 2009), S2a was not the dominant factor.

Another approach to qualitatively examine which organisms were responsible for controlling the distributions of concentrations of Hg in sediments is stepwise linear regression (Marvin-DiPasquale et al. 2009). Concentrations of Hg and S1, S2a, S2b, and RC were ln-transformed, and then a stepwise, the linear regression model was developed with the concentrations of Hg (as the dependent variable) and S1, S2a, S2b, and RC. In both QH and CH, concentrations of Hg in sediments were best described with a function that contained only S1, which accounted for 74 and 29 % of the variability, respectively (Fig. 5). The ratio of measured versus predicted concentrations of Hg in both lakes were approximately 1:1 (see Fig. 5). These results indicate that S1, which represents free hydrocarbons with smaller molecular weights in the SOM (Sanei et al. 2005; Sanei and Goodarzi 2006), played a major role in influencing Hg distributions in both lakes. Although the Hg concentrations of sediments in CH were also influenced by industrial development in southwestern China, it was still significantly controlled by S1.

Results observed in this study were consistent with those observed in eutrophic lakes at lower latitudes in central

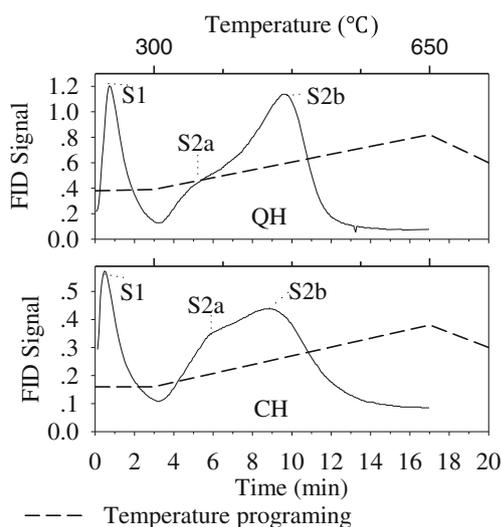
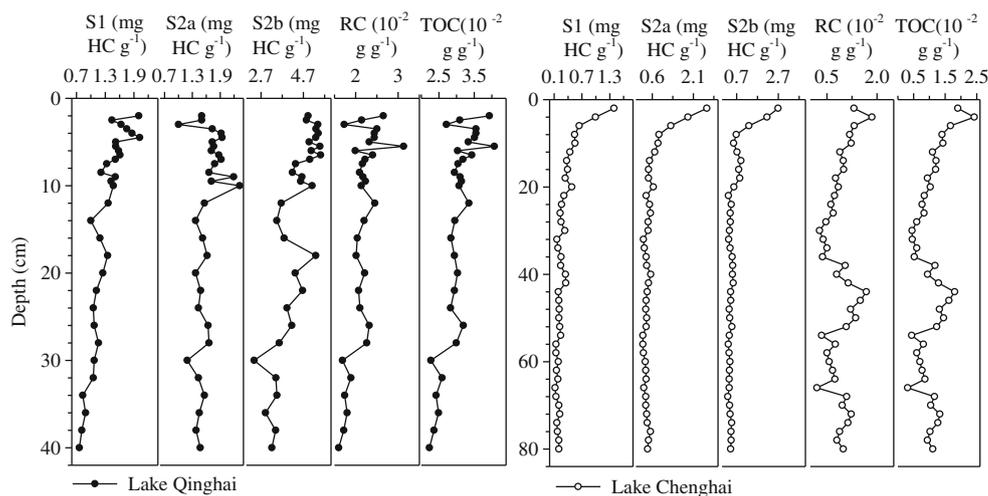


Fig. 3 Formation of hydrocarbon peaks (S1 and S2) during pyrolysis stage of sediment cores in Lakes Qinghai (QH) and Chenghai (CH)

Fig. 4 Vertical profiles of concentrations of S1, S2a, S2b, RC, and TOC in sediment cores from Lakes Qinghai and Chenghai



Alberta, Canada (Sanei and Goodarzi 2006), where SOM (S1) was the fraction of organic C that was most associated with concentrations of Hg in sediments. The authors of that study suggested that characteristics of SOM, including large surface area as well as chemical reactivity, were likely responsible for the association with concentrations of Hg and the reason for the scavenging of Hg from the water column. The results of the study reported here indicate that SOM plays a major role in Hg distributions in sediments not only of remote, less-polluted lakes (i.e., QH), but also of industrial-influenced, eutrophic lakes (i.e., CH).

Soluble organic matter might interact strongly with Hg and thus affect speciation, solubility, and mobility in the aquatic environment (Loux 1998; Ravichandran 2004; Wu et al. 2004; Fu et al. 2007; Bai et al. 2008). However, SOM from different sources might affect the dynamics of Hg differently (Ravichandran 2004). In order to further examine which fraction of DOM was most controlling of the dynamics of Hg distributions in sediments of CH and QH, profiles of HI and OI were compared with concentrations of Hg (Fig. 6). Land-derived terrestrial plants contain kerogen characterized by relatively greater OI than kerogen derived from algae, while algae contain kerogen characterized by relatively higher HI than land-derived terrestrial plants

(Stern et al. 2009). However, a recent study suggested that the HI could be influenced by terrestrial plants in the catchment such as conifer needles, so HI_a ((S2a/TOC)×100) was selected as the algal index (Carrie et al. 2012).

The profile of concentrations of HI_a in QH was different from the concentrations of Hg (see Fig. 6). HI_a exhibited an overall decreasing trend with decreasing depth, while Hg exhibited an overall increasing trend in QH. In CH, fluctuations in concentrations of HI_a were also different from those of Hg. The correlation coefficient between HI_a and Hg was 0.210 (*p*=0.193), which suggested that the correlation was not significant (see Fig. 6). Greater concentrations of HI_a suggest a greater proportion of organic C derived from algae. The fact that concentrations of Hg were not correlated with HI_a in the sediment core from CH and QH further suggests that organic C derived from algae was not the dominant factor controlling the distributions of Hg concentrations in sediments of CH and QH.

Different types of SOM exhibit different relationships with Hg concentrations. Relationships between concentrations of OI and Hg can also be used to understand the dynamics of Hg in lakes. In QH, trends in concentrations of OI as a function of depth in the sediment core were similar to those of Hg (see Fig. 6). From a depth of 40 to

Fig. 5 Measured versus predicted concentrations of Hg as a function of S1, determined by stepwise linear regression. Dashed diagonal lines represent the 1:1 line

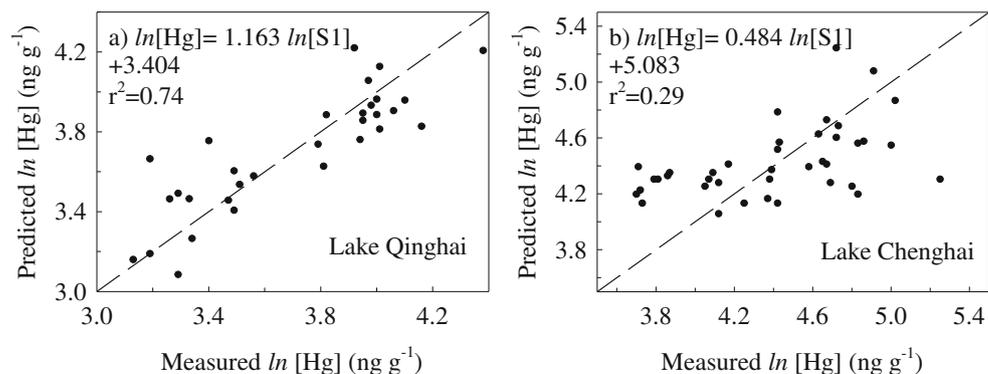
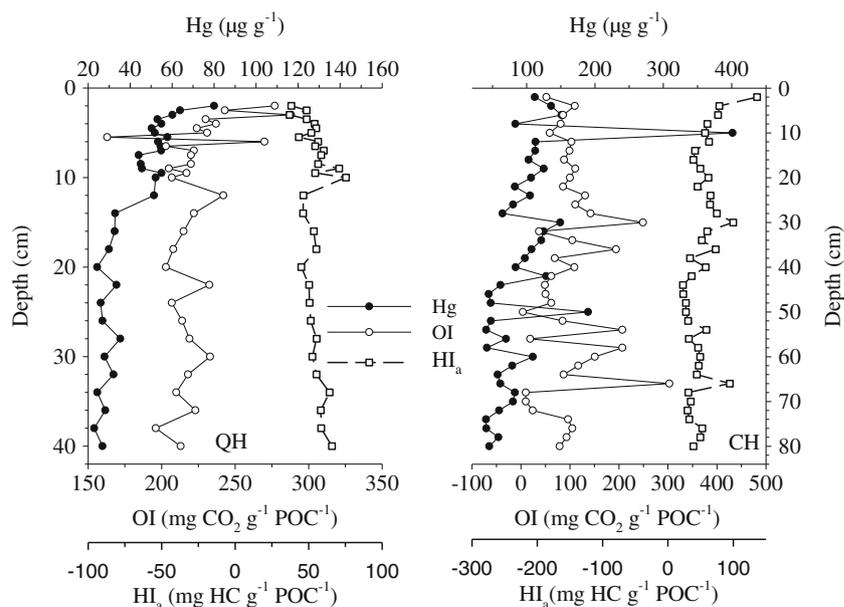


Fig. 6 Vertical profiles of concentrations of Hg, oxygen index (OI) and hydrogen index (HI_a) in sediment cores from Lakes Qinghai (QH) and Chenghai (CH)



32 cm and from 28 to 12 cm, trends in concentrations of OI were congruent with those of Hg; from a depth of 9.5 cm to the surface, OI had an overall increasing trend, which was similar to that of Hg (see Fig. 6). These results indicate that concentrations of Hg in sediments of QH were significantly influenced by allochthonous organic C derived from the terrestrial environment. Combined with the result of significant correlation between S1 and Hg observed in QH, it indicates that terrestrial SOM dominated distributions of Hg in sediments of QH. The strong binding of Hg by SOM is attributed to the coordination of Hg at reduced sulfur sites within the organic matter (Xia et al. 1999; Drexel et al. 2002; Ravichandran 2004). However, sulfur is a minor constituent of SOM, especially reduced sulfur which comprises the reactive functional groups expected to bind with Hg (Haitzer et al. 2002). Therefore, positive correlations between Hg and SOM could be expected in cases where Hg is primarily derived from wetlands and soils, where Hg has already combined with organic matter (Wallschläger et al. 1996; Ravichandran 2004). The catchment area of QH is six times greater than the surface area of the lake, and the main origin of Hg in the region is deposition from the atmosphere. Considering these factors together, it can be shown that the amount of Hg bound to OM derived from the terrestrial environment would be larger than that from direct deposition to the surface of the lake. Thus, S1 of terrestrial origin significantly controlled the distribution of Hg in sediments of QH. In recent years, the increasing population and also the development of agriculture and tourism in this area became greater, which has resulted in erosion of soils and increased the concentration of SOM of terrestrial origin entering QH. Greater concentrations of SOM from the terrestrial environment resulted in greater SOM combined with Hg entering QH. This might be

the reason that concentrations of Hg significantly increased in the core during recent years, from 3.5 cm to the top, for about 60 years (Xu et al. 2011a) (see Fig. 6).

In CH, the profile of concentrations of OI as a function of depth was different from that of Hg concentrations in the whole core. However, in the top 30-cm depth, fluctuations in concentrations were similar (see Fig. 6), except for the sudden increase of Hg concentrations at a depth of 10 cm, which might have been caused by an industrial event. This also indicates that, more recently, the distribution of Hg in sediments of CH was significantly influenced by SOM of terrestrial origin from the catchment, the area of which is almost three times greater than that of the lake surface.

The results for QH and CH provide further evidence that deposition of Hg in sediments could be significantly influenced by organic matter (Outridge et al. 2007; Stern et al. 2009). Results of studies of 14 Canadian Arctic and sub-Arctic lakes (Kirk et al. 2011) suggested that land-derived organic matter influenced a large portion of Hg concentrations in sediments and supported the result that land-derived SOM played an important role in sedimentary Hg distributions in this study. However, the conditions in the regions of China where these lakes are located are different from those in the Arctic (Outridge et al. 2007) and sub-Arctic lakes (Stern et al. 2009), the catchments of which consist of tundra that is covered most of the year by ice and snow, such that little SOM is washed into these Arctic lakes. Thus, SOM in Arctic and sub-Arctic lakes was primarily autochthonous, being derived from primary productivity within the lakes. Because S1 is the major organic fraction on the surfaces of sedimentary particles (Sanei et al. 2005; Sanei and Goodarzi 2006; Carrie et al. 2009), scavenging of Hg by algae might be mostly due to the binding of Hg to groups of S1 in the Arctic and sub-

Arctic lakes. This deduction is supported by the significant correlation between concentrations of S1 and Hg in the Arctic (Outridge et al. 2007) and sub-Arctic lakes (Stern et al. 2009). Therefore, SOM in the Arctic (Outridge et al. 2007) and sub-Arctic lakes (Stern et al. 2009) also dominated distributions of Hg in sediments, in a manner that is similar to the results for sediment cores from CH and QH and also eutrophic lakes of central Alberta, Canada (Sanei and Goodarzi 2006). Because SOM was derived primarily from primary productivity in the high latitude lakes, it is likely that scavenging of Hg by algae was the dominant mechanism that resulted in Hg being deposited in the sediments of those lakes (Outridge et al. 2007; Stern et al. 2009).

4 Conclusions

Results suggest that binding of Hg to SOM is one of the dominant processes which determine the depth distributions of Hg lake sediment cores. In lakes at lower latitudes, with large catchment areas that are not tundra, binding to allochthonous SOM is the dominant process controlling the distribution of Hg as a function of depth in sediments. Industries and other human activities, such as agriculture and tourism, can influence the distribution of Hg in lake sediments due to the greater erosion of soils by agriculture and tourism in those catchments. The results presented here provide further evidence that historical records of Hg in lake sediments might not only represent historical deposition of Hg directly to the lake, but are also significantly influenced by organic matter in the region.

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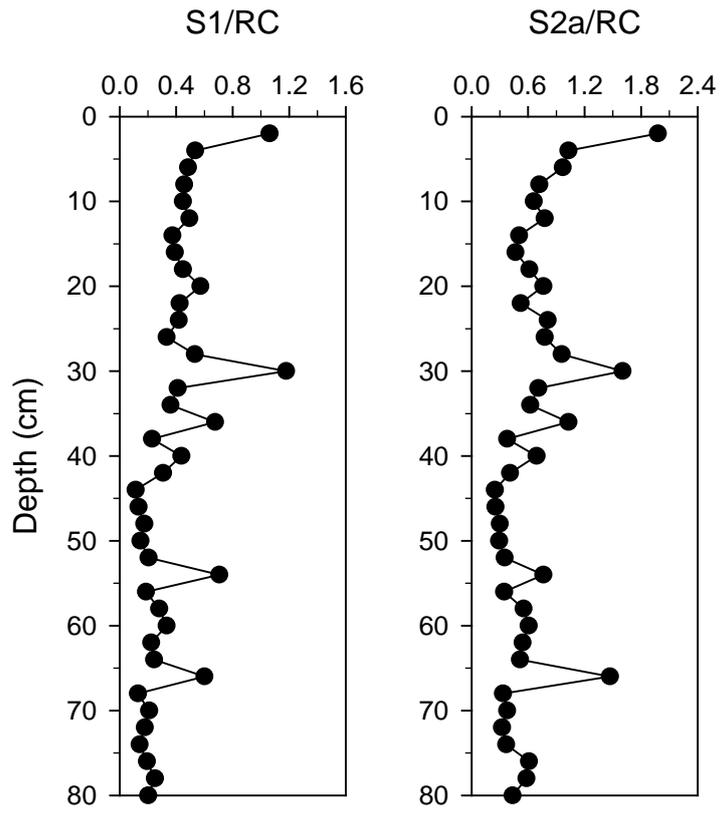


Fig. S1 S1/RC and S2/RC in sediment core of CH