Fractionation distribution and preliminary ecological risk assessment of As, Hg and Cd in ornithogenic sediments from the Ross Sea region, East Antarctica

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Abstract: To evaluate mobility of toxic elements and their potential ecological risk caused by seabird biovectors, the fractionation distributions of arsenic (As), mercury (Hg) and cadmium (Cd) were investigated in three ornithogenic sediment profiles from the Ross Sea region, East Antarctica. The results show residual As holds a dominant position, and Hg mainly derives from residual, organic matter-bound and humic acid-bound fractions, indicating weak mobility of As and Hg. However, exchangeable Cd occupies a considerable proportion in studied samples, suggesting Cd has strong mobility. The preliminary evaluation of Sediment Quality Guidelines (SGQs) shows adverse biological effects may occur occasionally for As and Cd, and rarely for Hg. Using Risk Assessment Code (RAC), the ecological risk is assessed at moderate, low and very high for As, Hg and Cd pollution, respectively. Organic matter derived from guano is the main factor controlling the mobility of Hg and Cd through adsorption and complexation.

Keywords: Arsenic; Mercury; Cadmium; Fractionation; Ornithogenic sediments, East Antarctica

1. Introduction

Seabirds act as biovectors between marine and terrestrial ecosystems, and are important material transport pathways in some remote coastal areas (Sun et al., 2000; Huang et al., 2014). The breeding activities of seabirds increase nutrients (such as nitrogen and phosphorus) into terrestrial habitats through excrement, and then the soil properties are modified to be more suitable for plant growth (Xu et al., 2011). In addition to the input of natural fertilizer, considerable toxic contaminants (such as heavy metals and persistent organic pollutants) are also transported by seabirds to their breeding sites, and may pose potential ecological risks to local ecosystems (Blais et al., 2005; Brimble et al., 2009; Michelutti et al., 2010). According to the results of our previous studies, the concentrations of As, Hg and Cd in ornithogenic sediments were strongly influenced by guano input in the Ross Sea region, East Antarctica (Nie et al., 2012; Liu et al., 2013), and intensive seabird activities could lead to remarkable enrichments of these elements in their neighboring soils and sediments. Besides the role of sink, contaminated sediments can also serve as a long-term source of heavy metals and metalloids because their stability and fate are strongly influenced by environmental conditions (Baig et al., 2009).

Fractionation distribution reflects the specific bound state of elements in soils and sediments, and induces a critical impact upon their mobility and bioavailability in the environment (Wenzel et al., 2001). Kumpiene et al. (2012) found that As preferentially associates with poorly crystalline Fe-oxyhydroxides in mine spoils, and the binding pattern did not inhibit As uptake by plants. Niazi et al. (2011) demonstrated that both exchangeable and amorphous Fe oxides-bound As in contaminated soils are readily available for plant uptake and may pose a potential risk to the environment. With respect to published studies on Hg fractionation, water-soluble and exchangeable Hg in polluted soils was highly correlated with gastrointestinal bioaccessibility, and these fractions could be potentially good indicators of Hg bioaccessibility (Zagury et al., 2006; Welfringer and Zagury, 2009). Similarly, significant correlations between Cd bioaccessibility and the sum of water-soluble and exchangeable Cd were found in incubated soil samples, and the results indicated that these forms of Cd were likely to constitute the main proportion of bioaccessible Cd in contaminated soils (Tang et al., 2006). Generally, fractionation

1 distribution of heavy metals in soils and sediments is influenced by complex factors
2 including grain size (Kocman et al., 2004), organic matter (Mohamed et al., 2010),
3 phosphate (Bolan et al., 2003), microbial behavior (Wang et al., 2012), and contamination
4 time (Lim et al., 2002). Up to now, most of the published studies on the fractionation of
5 heavy metals are concerned primarily with environmental pollution issues caused by
6 anthropogenic factors, such as contaminated sediments (Jain, 2004; Boughriet et al.,
7 2007) and mining soils (Varejão et al., 2009; Kreidie et al., 2011), and to our knowledge,
8 studies on heavy metal fractionation in natural ornithogenic sediments have not been
9 reported prior to now.

10 For example, As gradually accumulates in ornithogenic sediments with long-term
11 guano input, reaching high concentrations of up to 20 $\mu\text{g g}^{-1}$ (Liu et al., 2013). These
12 concentrations are much higher than the recommended safe level of As by SQGs, with a
13 value of 8.2 $\mu\text{g g}^{-1}$ in soils and sediments (Long et al., 1995), indicating that the exposed
14 ecosystem may suffer a potential risk of As pollution. However, the estimation of
15 ecological risk based only on total As levels may not be accurate in the region considered
16 in our study. Since mobility and bioavailability of As are dependent on its distribution
17 pattern among sediment partitions, fractionation analysis is more meaningful than the
18 estimation of its total concentration in assessing the sediment environmental quality.

19 Because of the remote location of Antarctica, direct human-induced disturbance is
20 insignificant. Therefore, it is beneficial for us to study the effect of seabird biovectors on
21 terrestrial environments under natural conditions. The primary objective of this study is to
22 analyze the fractionation distribution of As, Hg and Cd in ornithogenic sediments, and
23 thereby enhance our understanding about the environmental geochemistry behavior of
24 these heavy metals in seabird habitats. We also provide important implications for the
25 evaluation of ecological risk caused by penguin-derived potentially toxic elements
26 pollution in Antarctica.

2. Materials and methods

2.1 Sample collection

Three ornithogenic sediment profiles (MB6, BI and CC) and the fresh penguin

dropping sample (G1) were collected in the Ross Sea region, East Antarctica (Fig. 1). Among these, profiles MB6 and CC were collected at Cape Bird and Cape Crozier, respectively, and BI was at Beaufort Island. The detailed description of sampling sites and sectioning of the sediment profiles used in this study were reported by Nie et al. (2012). All the samples were kept at -20°C until in-depth studies were completed. Prior to chemical analysis, they were freeze-dried, homogenized and ground using an agate mortar and pestle, and then passed through a 200-mesh sieve.

2.2 Experimental section

In this study, six fractions of As, Hg and Cd, consisting of exchangeable fraction (including water-soluble fraction), carbonate-bound, humic acid-bound, iron/manganese (Fe/Mn) oxides-bound, organic matter-bound (including secondary sulfides) and residual fraction, were separated from ornithogenic sediment samples according to the analytical method reported by Liu et al. (2005). This experimental method is modified from Tessier et al. (1979), and the appropriate definition and optimized extraction procedure of different element species meet the demands of fractionation analysis in studies of environmental geochemistry. In the fractionation analysis experiment, subsamples of up to 2.5 g were placed in the centrifuge vessels, and the sequential extraction procedure for fraction analysis applied here (Table 1). The contents of As and Hg in detected sample solutions were analyzed by atomic fluorescence spectrometry (AFS), and Cd was analyzed by inductively coupled plasma-optical emission spectrometer (ICP-OES). All the measurements were determined at the Institute of Geological Experiment of Anhui Province. In the experimental procedures of fractionation analysis, two parallel experiments were conducted for all studied samples to confirm the measurement results. The detection limits of As, Hg and Cd in fractionation analysis are 0.10, 0.002 and 0.02 ($\mu\text{g g}^{-1}$), respectively. The relative standard deviation (RSD) of this method is better than 20%, and the extraction efficiency of six fractions is generally higher than 80% (Liu et al., 2005).

For phosphorus (P), 0.25 g subsamples were precisely weighed and acid digested ($\text{HNO}_3\text{-HF-HClO}_4$) in Teflon tubes with electric heating, followed by ICP-OES detection.

Total organic carbon (TOC) was measured by determining the samples' potassium dichromate oxidation capacity. Measurements of low frequency magnetic susceptibility (MS) were conducted using a Bartington MS2 susceptibility meter. For the profiles MB6 and BI, the grain size composition of subsamples was determined using the following treatment: 0.1 g sediment samples was air-dried before grinding and heated to 150°C with 10–20 mL H₂O₂ for 30 min, and then 10 mL HCl (10%) was added with heating to 80°C until no air bubbles were observed. After the removal of the supernatant following centrifugation, 10 mL deionized water was added and centrifugation was repeated. The samples were analyzed on a Laser Particle Size Analyzer (LS 13320) after 10 mL sodium hexametaphosphate (as dispersing agent) was added to the centrifuge tubes. Because of insufficient sample quantity, grain size analysis was not completed in the CC profile. Here, the definition of clay (<2 µm), silt (2–63 µm) and sand fraction (>63 µm) was based on the report of Holz et al. (2004).

3. Results and discussion

3.1 Fractionation distributions of As, Hg and Cd

The distribution of six As fractions in fresh guano (G1) and ornithogenic sediments (MB6, BI and CC) are given in Fig. 2. The results show that the residual fraction holds a dominant position in all measured samples with different As concentrations, and its content exhibits a similar trend with total As. The proportion of the residual fraction in the guano sample is 59.8%, while the average value is 61.4% in the ornithogenic sediments. In contrast, the concentration of the exchangeable fraction remains low. Exchangeable As content in the fresh guano is only 0.07 µg g⁻¹ (accounting for 4.3% of total As), and its average concentration in the ornithogenic sediments is 0.51 µg g⁻¹ (5.5% of total As). Of these, the highest level of exchangeable As in the ornithogenic sediments is 0.81 µg g⁻¹ in BI-6 (less than 10% of total As), indicating relatively weak mobility of As in the guano and ornithogenic sediments. In the fresh guano sample, the content of organic matter-bound As is 0.47 µg g⁻¹, and thus secondary in all six fractions (Fig. 2). Nevertheless, the situation is completely different in the ornithogenic sediments. The proportion of organic matter-bound As is negligible in these sediment samples, with the highest value being

only 2.2%, and its position is replaced by humic acid-bound, carbonate-bound and Fe/Mn oxides-bound As. The results show that the proportion of organic matter-bound As decreases significantly from fresh guano to ornithogenic sediments. After the excrement of seabirds is deposited, it goes through a long-term diagenetic process (Tatur et al., 1997). Thus, we suggest that the remarkable difference of As fractionation between guano and ornithogenic sediment samples may be related to diagenetic process of guano after deposition by seabirds.

The concentrations and proportions of Hg fractions in penguin guano and ornithogenic sediment are given in Fig. 3. The results show that Hg in the fresh guano is mainly composed of the residual fraction, organic matter-bound and humic acid-bound portion, accounting for 39.3%, 30.7% and 19.4% of total Hg, respectively. In the ornithogenic sediments, the main fraction components of Hg are similar to guano, although the specific values of each fraction differ in different samples. Exchangeable Hg content in fresh guano is 7.18 ng g^{-1} (nearly 10% of total Hg), and its average concentration and proportion in the ornithogenic sediments is only 1.71 ng g^{-1} and 2.43%, respectively. Thus, the mobility of Hg in these samples is rather weak, and it tends to be more stable in the diagenetic process of guano. The major difference of fractionation distribution between As and Hg is that the proportion of carbonate-bound and Fe/Mn oxides-bound Hg is minor in these sediment samples. Extremely high proportions of organic matter-bound and humic acid-bound Hg (exceeding 80% of total Hg) are noticeable in the upper layers of the MB6 profile, probably due to the sorption and complexation effects of organic matter in the sediments.

The results of Cd fractionation analysis in the guano and ornithogenic sediment samples are given in Fig. 4. Unlike As and Hg, exchangeable Cd occupies a dominant position in the fresh guano (52.8% of total Cd), while the proportion of organic matter-bound Cd is negligible. According to the fractionation results, we suggest that Cd in the guano sample has strong instability and mobility in the environment. Although this situation has been weakened in the ornithogenic sediments, the proportion of exchangeable Cd is still much higher compared with As and Hg, with a mean value of 24.5%. As a result, the proportions of carbonate-bound, humic acid-bound and Fe/Mn

oxides-bound Cd display varying degrees of increase in the ornithogenic sediments, indicating that the mobility of Cd decreases due to the adsorption and complexation effect by carbonates, humic acid and Fe/Mn oxides during the diagenetic process of guano. With the considerable input of guano with high Cd content, seabird habitat ecosystems are likely suffering from heavy Cd burden. At the same time of obtaining a lot of nutrients derived from guano, serious Cd pollution is probably a potential concern.

3.2 Effects of sediment characters on fractionation distribution

To discern possible controlling factors on the mobility of As, Hg and Cd in our sediments, physicochemical characteristics (including grain size, TOC and MS) of the ornithogenic sediment samples were determined (Table 2). The results show that the distribution of grain size in the MB6 profile is homogeneous, and the proportions of silt are all above 90% through the entire sediment profile. Similarly, data of median and average grain size in this profile does not display obvious fluctuation among the samples. However, the results of grain size analysis in the BI profile show that it can be clearly divided into three layers. Sand is predominant in the upper layers (> 70%), indicating relatively coarse grain size. In the middle layer, the distribution of grain size is similar to samples in the MB6 profile, and the silt fraction absolutely holds a dominant position. Sediment samples are mainly composed of silt and sand in the deeper layers, and each fraction accounts for nearly 50% of the total. Data of median and average grain size in the BI profile are accordant with the above results, and the grain size varies in the following order: the upper layer > the deeper layer > the middle layer. Referring to published literature, grain size may play an important role in the mobility and potential bioavailability of heavy metals in sediments. For example, Biester et al. (2004) suggested that the mobility of Hg in soils was inhibited with a high content of clay component. In contaminated soils by Hg mines, the contents of bioavailable Hg are positively correlated with the amount of clay and negatively correlated with the amount of sand, and this situation is opposite for the cinnabar fraction, showing positive correlations with sand and negative correlations with clay (Kocman et al., 2004). Heavy metals in exchangeable and carbonate fractions are weakly bound and sensitive to the external sedimentary environment change (Duan et al.,

2013). Pearson correlation analysis was determined between the mobility of heavy metals, expressed as the proportion of exchangeable fraction or the sum of exchangeable and carbonate fractions, and each grain size fraction in the MB6 and BI profiles, respectively. However, no obvious correlations were found in these sediment samples. We presumed that the possible reason was that the distribution of grain size in the sediments was very homogeneous, and one grain fraction (silt or sand) was always dominant in the specific sample. Thus, the composition of grain size appears to not be the main factor controlling the fractionation distributions of As, Hg and Cd in the ornithogenic sediments.

According to our previous studies, the ornithogenic sediments collected from the Ross Sea region were mainly composed of bedrock weathering materials and penguin droppings (Nie et al., 2012; Liu et al., 2013). The results of correlation analysis show that the mobility of As has no statistical correlation with both TOC and MS (Fig. 5). However, Hg and Cd in the three profiles exhibit a significantly negative correlation with TOC, and positive correlation with MS (Fig. 6), indicating that TOC and MS may be major factors controlling the mobility of Hg and Cd in these sediments. Biester et al. (2004) showed that Hg in contaminated soils by chlor-alkali plants was predominately bound to organic matter, and mainly sorbed onto mineral soil components only in sandy soils deficient in organic matter. Feyte et al. (2010) found that the anionic trace elements As and antimony (Sb) are preferentially bound to the authigenic Fe oxyhydroxides, whereas the other trace elements such as Hg and Cd are preferentially bound to the humic substances. In general, As and organic matter are negatively charged in soils and sediments, whereas Hg/Cd and Fe oxyhydroxides are positively charged. Thus, As tends to be adsorbed by Fe oxyhydroxides and Hg/Cd is more subject to the complexation effect of organic matter. According to the results mentioned above, we suggest that the difference in the nature of ionic charge between As and Hg/Cd can well explain their inconsistent correlation with TOC. Moreover, the significantly positive correlation between the mobility and MS for Hg and Cd is because the bedrock weathering materials (rich in inorganic mineral) and penguin guano (rich in organic matter) are the main components of the ornithogenic sediments, and this likely leads to opposite correlation results between MS and TOC. Based on our previous study and unpublished data, As speciation composition may

gradually experience the transformation from DMA (dimethylarsinate) to inorganic species in the diagenetic stage of guano (Lou et al., 2014). Because of different mobility between each As speciation, we presume this process may have some influence on the fractionation distribution of As in the ornithogenic sediments.

To determine the effect of guano input on the mobility of heavy metals, Cd is used as an example in the following discussion because of its high concentration and proportion of exchangeable Cd in the ornithogenic sediments. As a typical guano-derived element, the average content of P in the whole sediment profile is used to represent the degree of influence by seabird activity. The results show that the average values of total P content in the deeper layers of MB6 (No. 42-62) and the CC profile are significantly higher than those in the upper layers of MB6 (No. 5~10) and the BI profile (T-test, $p < 0.01$) (Fig. 7). The average values of humic acid-bound Cd and the sum proportion of humic acid-bound and organic matter-bound Cd in these profiles display similar distributions with total P, especially the average value of humic acid-bound Cd in the deeper layers of MB6, which is remarkably higher than other profiles (T-test, $p < 0.05$) (Fig. 7). However, the distributions of the exchangeable Cd proportion in these profiles are obviously inconsistent with total P; there is less mobility of Cd in the deeper layers of MB6 and the CC profile than in the upper layers of MB6 and the BI profile (Fig. 7). Although the penguin guano is the major source of Cd in the sediments (Liu et al., 2013), its mobility appears to be inhibited with substantial guano input, which is revealed by the fact that the exchangeable Cd proportion declines and humic acid-bound Cd proportion increases with the high P content in the ornithogenic sediments. In summary, the organic matter derived from guano in the ornithogenic sediments is the main factor controlling the mobility of Cd through adsorption and complexation.

3.3 Preliminary ecological risk assessment

SQGs have been widely used in ecological risk assessment of contaminated sediments. Chemical reagent concentrations, corresponding to the 10th and 50th percentiles of adverse biological effects, are called the Effects-range-low (ERL) and Effects-range-median (ERM), respectively. Sediments can be classified as either rarely

($<ERL$), occasionally ($\geq ERL$ and $<ERM$) or frequently ($\geq ERM$) associated with adverse biological effects (Christophoridis et al., 2009). The recommended ERL value for As is $8.2 \mu g g^{-1}$, and ERM is $70 \mu g g^{-1}$ (Long et al., 1995). Despite that total As levels in the sediment samples are all far less than the ERM value, 50%, 37.5% and 80% of samples in MB6, BI and CC profiles exceed the ERL value, respectively, indicating adverse biological effects may occur occasionally in these ornithogenic sediments as a result of As pollution. Because of the relatively weak stability of exchangeable and carbonates fractions, the sum of these two fractions is usually applied as the scale of the RAC, with the proportion below 1% being no risk, 1–10% being low risk, 11–30% being medium risk, 31–50% being high risk and above 50% being very high risk (Singh et al., 2005). The results show that the proportions of exchangeable and carbonate-bound As are between 2.8% and 24.1% (Fig. 2), with average values of 11.5%, 11.6% and 17.7% in MB6, BI and CC profiles, respectively, indicating that these sediments are at low to medium ecological risk caused by As pollution.

As for Hg, the recommended ERL and ERM values are $150 ng g^{-1}$ and $710 ng g^{-1}$, respectively (Long et al., 1995). Only in one sediment sample the total Hg level is slightly higher than the ERL value (BI-4, $155.08 ng g^{-1}$), showing that these ornithogenic sediments are rarely associated with adverse biological effects caused by Hg pollution. The results of RAC show that the proportions of exchangeable and carbonates fractions in all sediment samples are below 10%, even in two samples below 1% (Fig. 3). According to the definition of ecological risk level, 91.3% of sediment samples are at low ecological risk, and the remaining samples have no ecological risk caused by Hg pollution. Although the results of enrichment factor (EF) for Hg indicate apparent Hg accumulation in these sediment profiles caused by biotransportation (Nie et al., 2012), the situation of Hg pollution seems to be not enough to cause obvious ecological risks to local ecosystems.

The recommended ERL value for Cd is $1.2 \mu g g^{-1}$, and ERM is $9.6 \mu g g^{-1}$ (Long et al., 1995). Cd concentrations in 73.9% of sediment samples exceed the ERL value, indicating adverse biological effects probably occur occasionally in these ornithogenic sediments. The results of RAC show that the proportions of exchangeable and carbonates fractions in all sediment samples are between 36.1% and 75.6% (Fig. 4), and 47.8% of sediment

1 samples are at very high ecological risk caused by Cd pollution, with the other samples at
2 high risk level. In sum, seabird colonies are probably faced with a serious Cd pollution
3 threat because of high mobility and bioavailability of Cd in the guano and ornithogenic
4 sediment samples.
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10 **4. Conclusion**

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12 Residual As holds a dominant position in all tested samples, including guano and
13 ornithogenic sediments, indicating weak mobility of As. The proportions of exchangeable
14 and carbonate-bound As are between 2.8% and 24.1%, indicating that these sediments
15 are at low to medium ecological risk caused by As pollution. Moreover, the results of
16 SQGs indicate that adverse biological effects may occur occasionally in these
17 ornithogenic sediments as a result of As pollution. Hg in the fresh guano and ornithogenic
18 sediments is mainly composed of a residual fraction, organic matter-bound and humic
19 acid-bound portions, and these ornithogenic sediments are rarely associated with adverse
20 biological effects. The results of RAC show that most of these sediment samples are at
21 low ecological risk caused by Hg pollution. Unlike As and Hg, exchangeable Cd occupies
22 a considerable proportion in the fresh guano and ornithogenic sediments, indicating that
23 Cd has strong instability and mobility in the penguin-inhabited environment. The results of
24 RAC show that almost half of the samples are at very high ecological risk caused by Cd
25 pollution, with the other half at high risk level. For the factors controlling the As, Hg and Cd
26 mobility, no obvious correlations were found between their mobility and the grain size
27 compositions in all the sediment samples. Moreover, the mobility of As has no obvious
28 correlation with both TOC and MS, while Hg and Cd exhibit significantly negative
29 correlation with TOC, and positive correlation with MS. These results indicate that organic
30 matter content in the sediments is the controlling factor for the mobility of Hg and Cd
31 through adsorption and complexation. Since ornithogenic sediments are found globally
32 and often in coastal areas rich in biodiversity, this study will be beneficial for
33 understanding the fractionation distribution and potential ecological risk of seabird-derived
34 toxic elements in these regions, as well as in Antarctica.
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Fig. 1 Sampling sites in the Ross Sea region, East Antarctica.

Table. 1 Sequential extraction procedure for As, Hg and Cd fraction analysis used in this study. SSR and RT mean soil solution ratio and room temperature, respectively.

Sequence	Fraction	SSR	Extraction conditions
Step 1	Exchangeable fraction (including water-soluble fraction)	1:10	1 mol L ⁻¹ MgCl ₂ solution, shaking for 2 h at RT
Step 2	Carbonate-bound	1:10	1 mol L ⁻¹ NaAc solution, shaking for 5 h at RT
Step 3	Humic acid-bound	1:20	0.1 mol L ⁻¹ Na ₄ P ₂ O ₇ solution, shaking for 3 h at RT
Step 4	Fe/Mn oxides-bound	1:20	0.25 mol L ⁻¹ NH ₂ OH·HCl + 0.25 mol L ⁻¹ HCl solution, shaking for 6 h at RT
Step 5	Organic matter-bound (including secondary sulfides)	2.5:8	30% (v/v) H ₂ O ₂ + 0.02 mol L ⁻¹ HNO ₃ solution, water bath for 2.5 h at 83 °C
Step 6	Residual fraction	1:20	HCl + HNO ₃ + HClO ₄ , electric heating digestion

Fig. 2 Arsenic fractionation concentrations and proportions in the ornithogenic sediments from the Ross Sea region. G1 represents the fresh penguin guano sample, and MB6, BI and CC represent ornithogenic sediment samples.

Fig. 3 Mercury fractionation concentrations and proportions in the ornithogenic sediments from the Ross Sea region. G1 represents the fresh penguin guano sample, and MB6, BI and CC represent ornithogenic sediment samples.

Fig. 4 Cadmium fractionation concentrations and proportions in the ornithogenic sediments from the Ross Sea region. G1 represents the fresh penguin guano sample, and MB6, BI and CC represent ornithogenic sediment samples.

Table. 2 Physicochemical characteristics of the ornithogenic sediment samples.

	Clay (%)	Silt (%)	Sand (%)	Median grain size (μm)	Average grain size (μm)	TOC (%)	MS ($10^{-8} \text{ m}^3 \text{ kg}^{-1}$)
CC-4						4.98	295.73
CC-6						5.27	295.86
CC-8	–	–	–	–	–	5.07	230.75
CC-12						4.90	242.30
CC-16						3.96	1057.97
MB6-5	2.24	97.76	0	8.80	10.86	1.30	491.42
MB6-6	2.67	97.33	0	8.52	10.74	1.25	496.23
MB6-10	0.32	99.68	0	12.86	16.59	1.19	452.80
MB6-42	2.32	97.68	0	8.12	9.48	5.75	378.54
MB6-46	6.27	93.73	0	8.09	9.60	7.98	323.23
MB6-48	0	100	0	14.91	14.35	7.11	307.86
MB6-50	1.34	98.66	0	11.29	11.28	7.46	305.61
MB6-52	1.82	98.18	0	8.81	9.98	5.93	344.77
MB6-62	3.49	96.51	0	7.89	8.57	2.17	1010.63
BI-6	0.08	27.72	72.20	140.41	153.95	7.88	692.34
BI-9	0	13.14	86.86	327.36	320.36	5.57	712.82
BI-11	0.02	10.74	89.24	364.33	369.78	5.67	849.09
BI-27	0.01	99.99	0	26.57	22.24	3.95	876.79
BI-28	0.06	99.94	0	19.86	17.13	3.15	909.64
BI-29	0.24	99.76	0	19.74	15.67	3.24	968.83
BI-38	0	48.57	51.43	63.70	63.57	1.42	1580.53

Note: “–” means not determined.

Fig. 5 Pearson correlation analysis between the proportion of exchangeable As, the sum of exchangeable and carbonates As, TOC and MS in the sediment profiles of MB6, BI and CC.

Fig. 6 Pearson correlation analysis between the proportion of labile fractions of Hg and Cd, TOC and MS in the sediment profiles of MB6, BI and CC.

Fig. 7 Average values of total P, the proportion of exchangeable Cd, the proportion of humic acid-bound Cd and the sum proportion of humic acid-bound and organic matter-bound Cd in the upper and deeper sediment layers of MB6, BI and CC profiles.