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Mercury and methylmercury bioaccumulation in a contaminated bay

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ABSTRACT

The bioaccumulation and the main source of total Hg (THg) and methylmercury (MMHg) in the deposit-feeding polychaete *Neanthes japonica* collected in Jinzhou Bay, China, were investigated. Compared with the historical data, THg bioaccumulation in polychaetes collected in sediment of Jinzhou Bay was distinctly higher due to higher sediment THg concentration, but MMHg bioaccumulation was significantly lower. THg accumulation in polychaetes mainly derived from its accumulation in sediment. However, MMHg bioaccumulation in polychaetes did not correlate with Hg concentration in sediment. Besides sediment ingestion, MMHg accumulation in polychaetes may partially source from the process of in vivo transformation. The in vivo Hg methylation may take place in polychaetes, according to the excellent correlation between MMHg concentration and THg and inorganic Hg concentration in polychaetes. The biochemical characters in polychaete body, the oxidation-reduction environment and the microbial activity in polychaete gut may be beneficial to in vivo Hg methylation.

1. Introduction

Jinzhou Bay located at the northwestern bank of Bohai Sea in China is one of the most seriously heavy-metal polluted coastal areas, due to the anthropogenic pressures exerted by the activities of zinc smelting. The zinc smelter located at the coast of Jinzhou Bay is the largest zinc smelting plant in Asia. In addition, the non-ferrous metal smelting is significant anthropogenic mercury source (Wang et al., 2009). Mercury is well known to be a priority pollutant due to its persistence, high bioavailability and toxicity to organisms and human. Among mercury species, methylmercury (MMHg) is the most concern form because of its much higher toxicity, bioavailability and biomagnification. Total mercury (THg) and MMHg accumulation in coastal watersheds along the northwestern Bohai Sea coast substantially exceeded the background level and was comparable to that of the Hg mining area of China (Luo et al., 2012; Wang et al., 2009; Zheng et al., 2011). Although the metal pollution in Jinzhou Bay received intensive attention (Fan et al., 2014), few studies have systematically focused on Hg contamination in this extremely polluted Bay.

Mercury deposited in sediment can be converted to MMHg by a complex biogeochemical process under anoxic conditions. Methylmercury in sediment can be more readily bioaccumulated in deposit-feeding animals, biomagnified in food chains and produce adverse effect on human health. Previous studies have reported the elevated concentration of THg and MMHg in hydrophytes, aquatic animals and even in human hair in Huludao area of Liaoning Province, China (Wang et al., 2009; Zheng et al., 2011). The bioaccumulation of THg and MMHg associated with contaminated sediment in the base of food chain is vital to understand Hg bioavailability and toxicity to the higher trophic level organisms. The deposit-feeding polychaetes are dietary item for several estuarine predators (e.g. fish, crabs and birds), and they are important vector for Hg biomagnification in the estuarine food chain (Coelho et al., 2008; Sizmur et al., 2013a). Therefore, Hg bioaccumulation in polychaetes is of great significance to understand the harm of Hg to ecological environment and human health.

The source of Hg bioaccumulation in organisms could influence the prediction of Hg trophic transfer and biomagnification in aquatic food chains. Generally considering, Hg and MMHg accumulation in deposit-feeding organisms mainly sourced from sediment (Wang et al., 1998). Additionally, MMHg accumulated in organisms may be also produced by the organisms themselves involving coenzymes or the microbial activities in their body or gut (Ridley et al., 1977). The transformation

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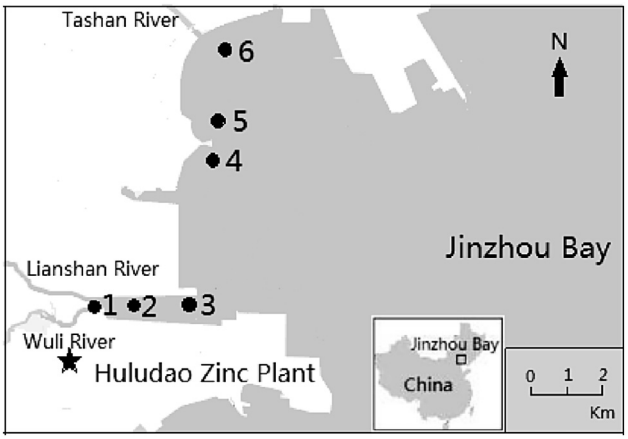

Fig. 1. Map of sampling locations in Jinzhou bay (CHINA).

of inorganic Hg to MMHg has been found in vivo in earthworm, fish and aquatic macrophyte (Göthberg and Greger, 2006; Rieder et al., 2013; Wang et al., 2013). However, the possible source and production mechanism of MMHg accumulated in benthic invertebrates remain poorly investigated.

In this study, we collected sediment and polychaetes *Neanthes japonica* from different sites in Jinzhou Bay with strong environmental gradients of sediment contamination. The accumulation of THg and MMHg in sediment and polychaetes was investigated. Moreover, the correlations of THg and MMHg bioaccumulation in polychaetes with their accumulation in sediment were analyzed. The source of THg and MMHg accumulated in polychaetes was discussed. The possible mechanism of MMHg production in polychaetes was considered.

2. Materials and methods

2.1. Sampling

Samples of sediments and polychaetes (*N. japonica*) were collected from 6 different sites of intertidal areas in Jinzhou Bay, as shown in Fig. 1. Samples were taken in each site at the low tide in July 2009. The sediment samples were collected from the upper 1–2 cm with a plastic grab, placed into polyethylene bottles and then immediately sealed in bags. The polychaetes were rinsed from mucus and sediment with seawater, and depurated to purge the sediment particles in the guts of polychaetes for 24 h after collection. The sediments and depurated polychaetes were subsequently placed in a cool-box and immediately transported to laboratory. Then, the sediments and polychaetes were stored at −80 °C until further processing.

2.2. THg and MMHg concentration in sediment and polychaetes

To measure THg and MMHg concentration in samples, sediment and polychaetes were dried at 50 °C for 3 days. The dried sediments were ground in a mortar and then sieved through 63 μm mesh to ensure the consistent physical properties. THg in sediment and polychaetes was measured using cold vapor atomic fluorescence spectrometry (CVAFS) after digested by acids (1,3 HCl + HNO_3) at 95 °C (Feng et al., 2009; Qiu et al., 2006). MMHg in sediment and polychaetes was determined by gas chromatography-CVAFS after solvent extraction using HNO_3 leaching/CH_2Cl_2 followed by ethylation onto Tenax traps as described in (Liang et al., 2004; Liu et al., 2012). THg and MMHg concentrations were expressed on dry weight and wet weight bases in sediment and polychaetes, respectively. The detection limits of the method were 10 μg/kg dry wt for THg and 0.005 μg/kg dry wt for MMHg. The analytical accuracy of THg and MMHg in sediment was checked by simultaneous digestion and analysis of standard sediment (IAEA-158, Marine Sediment). The recoveries of THg and MMHg were calculated as the ratio of the measured concentration of THg and MMHg divided by the concentration of THg and MMHg in the standard reference materials. Recoveries were 85–116% for THg and MMHg. The quality controls of THg and MMHg in polychaetes were conducted by simultaneous digestion and analysis of certified reference material IAEA-142 (mussel homogenate), with the recovery of 91–106% for THg and MMHg. In addition, the concentration of inorganic Hg was calculated by subtracting the concentration of MMHg from THg.

2.3. Data analysis

Any change of THg and MMHg concentration in sediment and bioaccumulation in polychaetes was tested using one-way analysis of variance (ANOVA). Post hoc tests were applied to identify differences between groups ($p < 0.05$). Linear-regression analysis was used to determine the regression coefficient (r^2) and the significance of the linear relationships ($p < 0.05$).

3. Results and discussion

3.1. THg and MMHg concentration in sediments

THg and MMHg concentrations in sediments collected from 6 sites of Jinzhou Bay were shown in Fig. 2. A wide range of THg concentrations from 432 μg/kg to 11,885 μg/kg was observed in sediment. Concentrations of MMHg varied from a minimum of 0.67 μg/kg to 8.56 μg/kg. Obviously, THg and MMHg concentrations were significantly higher in the sediment of site 2 collected near the Zn smelting factory than those in other stations. Zn smelting operation played a dominant role in THg and MMHg pollution in Jinzhou Bay.

The highest THg concentration in sediment of Jinzhou Bay was about 339 times of the background value in the sediment of Liaoning Province (35 μg/kg) (Sun, 1992) and 59 times higher than the China national guideline value (200 μg/kg, GB 18668–2002). THg and MMHg concentrations in sediment of Jinzhou Bay were significantly higher than those in the sediment collected from other coastal areas of China and other countries (e.g., THg: 7–398 μg/kg in the four Chinese marginal seas (Bohai Sea, Yellow Sea, East China Sea and South China Sea), MMHg: 0.006–0.098 μg/kg in the East China Sea, THg: 5.8–225 μg/kg and MMHg: 0.061–0.94 μg/kg in the Southern Baltic, THg: 12–90 μg/kg and MMHg: 0.014–1.5 μg/kg in the Mekong Delta, THg: 20–2400 μg/kg in the Kedougou region of the eastern Senegal, THg: 10–200 μg/kg and MMHg: 0.02–5 μg/kg in South Africa, THg: < 20 μg/kg and MMHg: 0.1–0.5 μg/kg in Canada, THg: 0.62–68.8 μg/kg and MMHg:

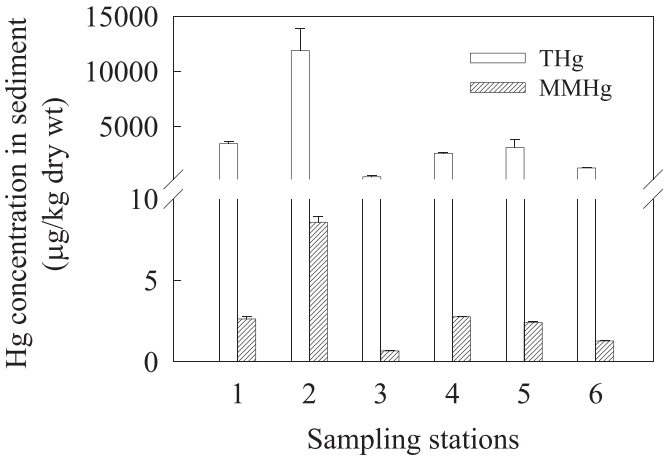

Fig. 2. The concentration of total mercury (THg) and methylmercury (MMHg) in sediment. Data are mean ± SD (n = 3).

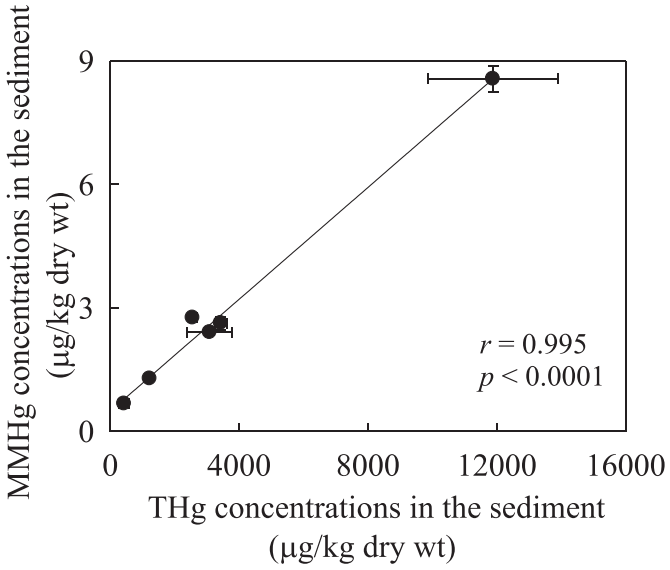

Fig. 3. The correlation of MMHg concentration in sediment with THg concentration in sediment. Data are mean ± SD (n = 3).

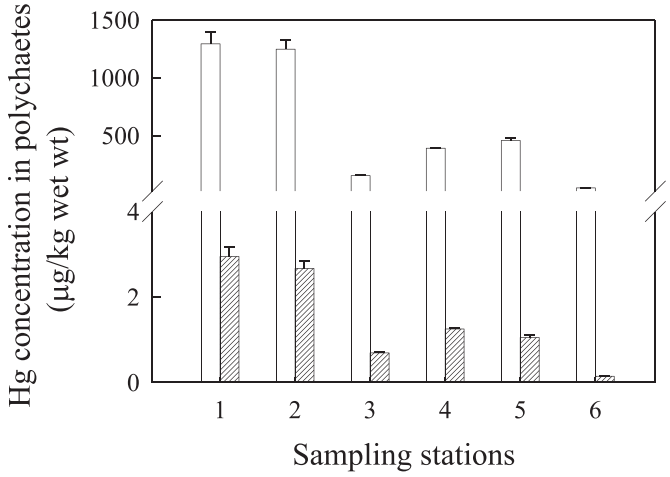

Fig. 4. The bioaccumulation of THg and MMHg in polychaetes. Data are mean ± SD (n = 3, each had a composite of 10–20 worms).

0.008–0.96 μg/kg in the mid-Atlantic continental shelf and slope) (Bełdowski et al., 2014; Hollweg et al., 2010; Li et al., 2018; Meng et al., 2014; Niane et al., 2019; Sizmur et al., 2013a; Walters et al., 2011; Zhao et al., 2019). THg and MMHg concentrations were comparable with those in the sediment of adjacent Marano and Grado Lagoon in Italy (THg: 680–9950 μg/kg and MMHg: 0.47–7.85 μg/kg), the northwest coast of Portugal (THg: 200–11,900 μg/kg) (Acquavita et al., 2012; Nunes et al., 2008) and the Kedougou region of the eastern Senegal (MMHg: 2.3–8.0 μg/kg) (Niane et al., 2019), which had also been seriously contaminated by anthropogenic activity.

The concentrations of THg in sediment of Jinzhou Bay correlated strongly with MMHg concentration in sediment ($r = 0.995$, $p < 0.0001$), as shown in Fig. 3. It indicated that THg concentration considerably affected the methylation rate of Hg and controlled MMHg concentration in sediment. THg was of importance for the long-term accumulation of MMHg in sediment (Drott et al., 2008). Previous study also reported that MMHg concentration in sediment showed a significant relationship with the concentration of labile Hg and THg (Conaway et al., 2003; Sizmur et al., 2013a; Wang et al., 2009). The dominant forms of Hg in aquatic environment are the inorganic Hg and MMHg. MMHg in sediment of Jinzhou Bay may source from the transformation of inorganic Hg besides anthropogenic activity.

The biogeochemical factors including physicochemical variables of environment and sediment, Hg speciation and the anaerobic microbial community in sediments govern MMHg production (Buckman et al., 2019; Jonsson et al., 2014; Ndu et al., 2018; Zhang et al., 2014; Zhao et al., 2019). The environmental conditions (temperature, salinity and seawater) may change Hg distribution and in sediment (Buckman et al., 2019; Chakraborty et al., 2019; Zhao et al., 2019). Hg preferred to associate with the smallest grain size sediment fractions with high total organic carbon content (Chakraborty et al., 2014; Reinhart et al., 2018). The nature and source of sedimentary organic matter played a crucial role in Hg speciation and MMHg formation (Buckman et al., 2019; Chakraborty et al., 2015; Zhao et al., 2019). Phytoplankton-derived organic compounds in sediment could increase mercury methylation rates with increasing bacterial activity (Bravo et al., 2017). In addition, organic matter may also affect the microbial availability of different Hg species and thus MMHg production in sediment (He et al., 2019).

The inorganic Hg^{2+} in sediment usually combined with geochemical phases and presented different chemical species. The chemical species of Hg^{2+} in sediment significantly influenced Hg methylation. The neutral inorganic mercury sulfide species and low molecular-mass Hg thiols with high abundance of natural organic matters may be available for Hg methylation and control Hg methylation rate in the contaminated sediment (Drott et al., 2007; Jonsson et al., 2014). Liem-Nguyen et al. (2016) also reported some Hg species, such as Hg-S complexes and low-molecular-weight Hg-thiol complexes, were more bioavailable to microbial methylators in favor of MMHg production.

In addition, sulfate reducing bacteria (SRB), iron reducing bacteria and methanogens can cause Hg methylation in sediment (Erickson and Lin, 2015). The first dominant bacterial community in Bohai Sea intertidal sediment was *Proteobacteria*, which was classified belonged to *Deltaproteobacteria* (Zheng et al., 2014). Most of the *Deltaproteobacteria*-like sequences correlated with the sulfate-reducing and iron-reducing bacteria (Liu et al., 2014). It provided evidence that there were the *Hg*-methylating bacteria living in intertidal sediment of Bohai Sea, which played a vital role in the transformation of Hg to MMHg.

3.2. THg and MMHg in polychaetes

Tissue concentrations of THg and MMHg in polychaetes collected from the sediment in Jinzhou Bay were analyzed (Fig. 4). THg and MMHg concentrations in polychaetes were 47–1294 μg/kg and 0.15–2.95 μg/kg, respectively. THg and MMHg accumulation in polychaetes was substantially higher at the polluted sites (sites 1 and 2) near the Zn smelting factory, in accordance with THg and MMHg pollution in the sediment of Jinzhou Bay.

We compared THg and MMHg bioaccumulation in polychaetes collected from the sediment in Jinzhou Bay with those in previous reports (e.g. THg: 23.2–434 μg/kg and MMHg: 2.88–69.6 μg/kg in polychaetes collected from the Bay of Fundy in Canada; THg: 39–130 μg/kg and organic Hg: 1.33–56 μg/kg in the ragworm *Hediste diversicolor* (Coelho et al., 2008; Sizmur et al., 2013a)). It was found that THg bioaccumulation in polychaetes collected in sediment of Jinzhou Bay was extremely higher, but MMHg bioaccumulation was relatively lower. The extremely higher THg bioaccumulation in this study may be due to the quite higher THg concentration or bioavailable Hg partition in sediment of Jinzhou Bay than those measured in previous studies.

MMHg bioaccumulation may be affected by many environmental factors (e.g., sediment and water salinity, water MMHg concentration, water dissolved organic carbon and local landscape variables) (Buckman et al., 2017; Chételat et al., 2018; Reinhart et al., 2018). Buckman et al. (2017) found in their study of MMHg bioaccumulation in fauna from the Delaware River estuary that drivers of MMHg bioavailability were complex and MMHg bioaccumulation was driven by a combination of water MMHg concentration and local landscape

characteristics (human development, marsh cover, forest cover, salinity and total suspended solids). The difference of these environmental factors in different regions may result in the difference of MMHg bioaccumulation between this study and previous studies.

In addition, the differences of family, feeding ecology and physiological traits for polychaetes between this study and previous studies may be another influence factor. The polychaetes (*N. japonica*) in this study from the family of nereididae are described as surface deposit-feeders and omnivores (Kikuchi, 1987). However, the polychaetes with higher MMHg bioaccumulation are from the family of maldanidae in the Bay of Fundy in Canada and usually behave as subsurface and deep burrowing deposit-feeders (Sizmur et al., 2013a). The feeding depth of polychaetes influence Hg bioaccumulation and the polychaetes feeding on deeper sediments contained greater MMHg concentrations (Sizmur et al., 2013a). The higher MMHg dietary opportunism of the polychaetes in previous studies may be the reason for the elevated MMHg bioaccumulation. The ragworm *H. diversicolor* (14 mg dry weight for adults) in the northwestern coast of Portugal of previous studies is larger than the polychaetes *N. japonica* (3 mg dry weight for adults) in this study, although it is from the same family of nereididae as *N. japonica* (Rosen and Miller, 2011). In addition, different from the polychaetes *N. japonica* in this study, the ragworm *H. diversicolor* does not build a mucus-lined tube, which can potentially increase the exposure to sediment contaminants. These different physiological traits of polychaetes may result in different MMHg bioaccumulation. Besides, the source of MMHg in organisms may be another factor affecting MMHg bioaccumulation.

3.3. The source of THg and MMHg in polychaetes

There was positive relationship between THg accumulation in polychaetes and THg concentration in sediment ($r = 0.764$, $p < 0.05$), as seen in Fig. 5(a). The body burden of THg in polychaetes was mainly through ingested sediment. Wang et al. (1998) also predicted by bioenergetic-based kinetic model that > 70% Hg accumulation in polychaetes was from sediment ingestion (Wang et al., 1998).

MMHg concentration in polychaetes did not correlate with THg ($r = 0.715$, $p = 0.07$), MMHg ($r = 0.706$, $p = 0.08$) and inorganic Hg concentrations in sediment ($r = 0.684$, $p = 0.134$), as shown in Fig. 5(b), (c) and (d). It implied that MMHg concentration in polychaetes was not closely affected by Hg concentrations in sediment. The same results were also observed in the relationships between MMHg concentrations in sediment and invertebrates (e.g., chironomids, polychaetes, crabs and grass shrimp) collected near the Bay of Fundy in Canada and the Delaware River estuary in northeastern USA (Reinhart et al., 2018; Sizmur et al., 2013a). Sediment Hg concentrations were not a good predictor of MMHg bioaccumulation. As mentioned above, MMHg bioaccumulation could not be predicted by one single variable and was driven by a combination of environmental factors.

MMHg concentration in polychaetes significantly correlated with the concentration of THg ($r = 0.978$, $p < 0.0001$) and inorganic Hg ($r = 0.989$, $p < 0.0005$) in polychaetes, as plotted in Fig. 5(e) and (f). Such correlation appeared to be much better than that of MMHg concentration in polychaetes relating with THg and MMHg concentrations in sediment. These data suggested that THg and inorganic Hg concentrations in polychaetes controlled MMHg bioaccumulation. A portion of MMHg accumulation in polychaetes may source from the process of in vivo transformation besides sediment ingestion. Inorganic Hg may be transformed into MMHg in the body of polychaetes. The in vivo methylation of Hg may take place in polychaetes.

In addition, the ratio of MMHg to THg (%MMHg) in polychaetes (averaged 0.29) was much greater than that in sediment (averaged 0.098). Similarly, the biota-sediment accumulation factors (BSAF) values of MMHg (0.11–1.12) were obviously higher than those of THg (0.04–0.38) (Table 1). BSAF higher than one was found for MMHg in polychaetes collected from sites close to the factory. These results illustrated that MMHg can be more easily biomagnified in the aquatic food chain than inorganic Hg. The assimilation efficiency of MMHg (43–83%) in polychaetes was significantly higher than that of inorganic Hg (7–30%) (Wang et al., 1998). On the other hand, the transformation of Hg into MMHg in polychaetes may occur and play a role in the increase of %MMHg and BSAF of MMHg in polychaetes at some extent. Previous study, which showed earthworms were potentially able to methylate Hg, also found %MMHg was much higher in earthworm tissue than in the substrate (Hinton and Veiga, 2002).

The speciation of Hg(II) in sediment controlled the bioavailability of Hg(II). The Hg(II) in surface sediment primarily distributed in the organocomplexed (e.g., Hg humic and Hg_2Cl_2) and strong-complexed (e.g., Hg bound up in Fe/Mn oxide, amorphous organosulfur, or mineral lattice) geochemical phases, quantified by sequential chemical extraction method (Yu et al., 2012). Most Hg(II) bound with the strongly complexed geochemical phases was bioavailable, whereas Hg came from organocomplexed phase was not bioavailable in the benthic invertebrates clam *Ruditapes philippinarum* and the gut juices extraction from the sipunculan *Sipunculus nudus* (Zhong and Wang, 2006). Therefore, Hg(II) bound with the strongly complexed geochemical phases in sediment may be bioavailable and assimilated in polychaetes. Hg-sulfide complexes and low-molecular-mass Hg-thiol complexes were more bioavailable to microorganisms methylating Hg to MMHg (He et al., 2019; Liem-Nguyen et al., 2016). Hg(II) originated from these strongly complexed geochemical phases in sediment ingested by polychaetes may participate in the transformation of Hg into MMHg in polychaetes.

Although there has been no relevant report about the in vivo methylation in benthic invertebrates, the biochemical characters in the body of the polychaetes may be beneficial to the transformation of inorganic Hg to MMHg. Sizmur et al. (2013) found the mucus secretions and organic detritus in polychaetes increased the concentration of MMHg and liable Hg(II) in sediment (Sizmur et al., 2013b). In addition, there were a large number of aerobic and anaerobic microorganisms, such as methanogens and SRB, in the gastrointestinal tracts of polychaetes (Li et al., 2009). The methanogens and SRB was the main microorganisms methylating Hg to MMHg. Therefore, the microorganisms in the gut of polychaetes may play an important role in the in vivo Hg methylation. Moreover, the oxygen concentration and the oxidation-reduction potential in polychaete gut presented an environment gradient (Li et al., 2009). The shifting of redox conditions could enhance the microbial activity and further promote the formation of MMHg (Canário et al., 2007). Therefore, the oxidation-reduction environment of polychaete gut was also beneficial to the transformation of Hg to MMHg. The production of MMHg in the body of benthic invertebrates was firstly put forward in this study. The mechanism of the in vivo Hg methylation in polychaetes should be further confirmed.

In vivo methylation has been found in earthworm and aquatic biota such as the aquatic macrophyte and fish (Cosio et al., 2014; Rieder et al., 2013; Wang et al., 2013). The potential of in vivo methylation was very low (0.67–1.60%) in freshwater tilapia (*Oreochromis niloticus*) (Wang et al., 2013). However, the potential of in vivo methylation reached as high as about 85% in earthworm (Rieder et al., 2013). The in vivo transformation of inorganic Hg to MMHg in polychaetes has not been reported. The potential of in vivo Hg methylation in polychaetes is interesting and meaningful to understand the speciation and final biological fate of Hg bioaccumulated in polychaetes. It was worthy to be further investigated.

4. Conclusions

Compared with other coastal areas of China and other countries, THg and MMHg concentrations in sediment of Jinzhou Bay were much higher than sediment background value and guideline value. Due to higher sediment THg concentration, THg bioaccumulation in polychaetes collected in sediment of Jinzhou Bay was distinctly higher

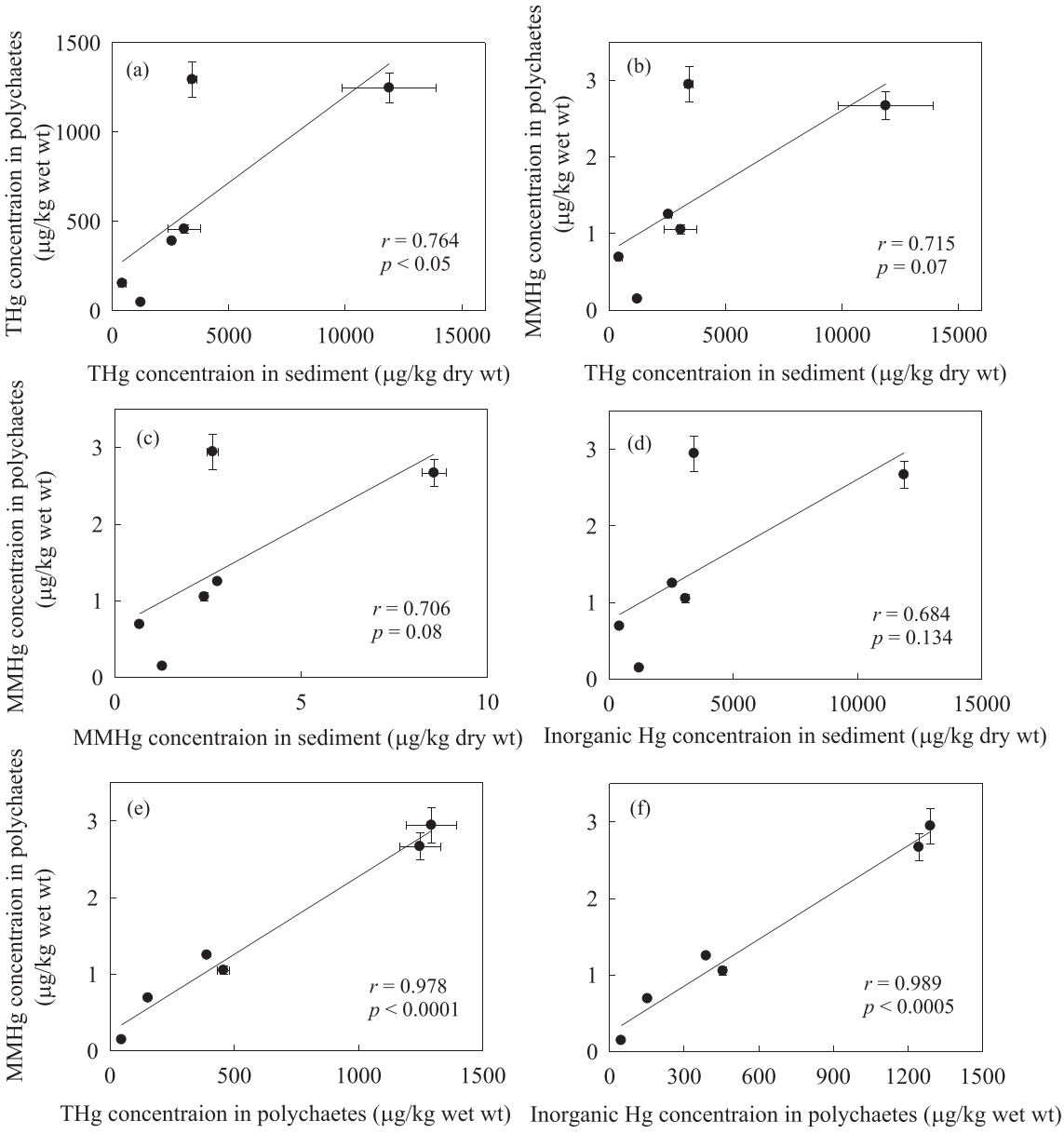

Fig. 5. The relationships of THg, MMHg and inorganic Hg concentration between in sediment and polychaetes (a) THg in sediment and polychaetes (b) THg in sediment and MMHg in polychaetes (c) MMHg in sediment and in polychaetes (d) inorganic Hg in sediment and MMHg in polychaetes (e) THg and MMHg in polychaetes (f) inorganic Hg and MMHg in polychaetes. Data are mean ± SD (n = 3, each had a composite of 10–20 worms).

Table 1
The biota-sediment accumulation factors (BSAF) of THg and MMHg in polychaetes and the ratio of MMHg to THg (%MMHg) in sediment and polychaetes.

Stations	$BSAF_{THg}$	$BSAF_{MMHg}$	%MMHg in sediment	%MMHg in polychaetes
1	0.38	1.12	0.08	0.23
2	0.10	0.31	0.07	0.21
3	0.35	1.03	0.15	0.45
4	0.15	0.45	0.11	0.32
5	0.15	0.44	0.08	0.23
6	0.04	0.11	0.10	0.31

relative to that in previous reports. However, MMHg bioaccumulation was significantly lower. THg accumulation in polychaetes mainly derived from its accumulation in sediment. However, MMHg accumulation in polychaetes was not correlated with Hg concentration in sediment. The excellent correlations of MMHg bioaccumulation with THg and inorganic Hg concentration in polychaetes indicated that MMHg production in the body of polychaetes might occur. MMHg accumulation in polychaetes may partially source from the in vivo Hg methylation. The biochemical characters in polychaete body, the oxidation-reduction environment and the microbial activity in the gastrointestinal tracts of polychaetes may play an important role in the in vivo transformation of Hg to MMHg. The mechanism and potential of the in vivo Hg methylation in polychaetes should be further confirmed.

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