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Methylmercury and sulfate-reducing bacteria in mangrove sediments from Jiulong River Estuary, China

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Abstract

Estuaries are important sites for mercury (Hg) methylation, with sulfate-reducing bacteria (SRB) thought to be the main Hg methylators. Distributions of total mercury (THg) and methylmercury (MeHg) in mangrove sediment and sediment core from Jiulong River Estuary Provincial Mangrove Reserve, China were determined and the possible mechanisms of Hg methylation and their controlling factors in mangrove sediments were investigated. Microbiological and geochemical parameters were also determined. Results showed that SRB constitute a small fraction of total bacteria (TB) in both surface sediments and the profile of sediments. The content of THg, MeHg, TB, and SRB were (350 ± 150) ng/g, (0.47 ± 0.11) ng/g, ($1.4 \times 10^{11} \pm 4.1 \times 10^{9}$) cfu/g dry weight (dw), and ($5.0 \times 10^{6} \pm 2.7 \times 10^{6}$) cfu/g dw in surficial sediments, respectively, and (240 ± 24) ng/g, (0.30 ± 0.15) ng/g, ($1.9 \times 10^{11} \pm 4.2 \times 10^{10}$) cfu/g dw, and ($1.3 \times 10^{6} \pm 2.0 \times 10^{6}$) cfu/g dw in sediment core, respectively. Results showed that THg, MeHg, TB, MeHg/THg, salinity and total sulfur (TS) increased with depth, but total organic matter (TOM), SRB, and pH decreased with depth. Concentrations of MeHg in sediments showed significant positive correlation with THg, salinity, TS, and MeHg/THg, and significant negative correlation with SRB, TOM, and pH. It was concluded that other microbes, rather than SRB, may also act as main Hg methylators in mangrove sediments.

Key words: mercury; methylmercury; sediment; mangrove; sulfate-reducing bacteria

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Introduction

Mercury (Hg) is a significant contaminant in estuary environments, particularly as methylmercury (MeHg) forms in aquatic sediments and bioaccumulates in food webs (Heyes et al., 2006). Even at trace quantities in water or sediment, MeHg shows a marked tendency to bioaccumulate in organisms, particularly fish species (Ouddane et al., 2008). Methylation of Hg in the aquatic environment has been considered largely the result of biological processes involving the activities of sulfate-reducing bacteria (SRB) (Compeau and Bartha, 1985; Hall et al., 2008). Through diffusion and remobilization from surface sediments, a fraction of MeHg periodically transfers into the water column (Heyes et al., 2006; Hammerschmidt and Fitzgerald, 2006; Castelle et al., 2007). It is these processes that play a key role in MeHg bioaccumulation in estuarine ecosystems.

Although understanding the fate of Hg in near-coastal ecosystems has been a research focus for many years, information is still less available than that for freshwater aquatic systems. Wetlands are of major concern in environmental science as they have been identified as important sites for Hg methylation (Gilmour and Henry, 1991). Due to large productivity and biodiversity, mangrove wetlands are important ecosystems in both tropical and subtropical areas with highly complex food webs. Through biological cycles and exchanges between sediment and water, MeHg enters the food chain and is of concern in regards to public health. Mangrove wetlands possess ideal characteristics for studying SRB and Hg methylation, including high sulfate content, high organic matter content, acidity, high salinity, and low oxygen.

This study was conducted in Jiulong River Mangrove Provincial Reserve (JRM) (24°20′–24°32′N, 117°54′–118°03′E). Jiulong River is the second biggest river in Fujian Province, China, with a mean annual temperature of 21.0 and a mean annual rainfall of 1365 mm. On the

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north verge of a natural mangrove, JRM has an area of 87.4 ha covered by mangroves (Lin, 1997) in which *Kandelia candel*, *Avicennia marina*, and *Aegiceras corniculatum* occur. Various studies have been conducted in JRM since the 1980s, including the distribution of organochlorine pesticides (Lin and Huang, 1994), the cycling of heavy metals (Zheng et al., 1996a, 1996b, 1996c), and the source, distribution, and degradation of PAHs in mangrove ecosystems (Tian et al., 2008a, 2008b). Except for a brief report on total mercury (THg) distribution (Ding et al., 2009), little information is available on biogeochemical cycling of Hg in this mangrove ecosystem.

The aim of this study was to examine THg and MeHg distribution in sediments, especially in the sediment profile of this mangrove wetland. We explored correlations among MeHg concentrations and environmental factors such as pH, salinity, total organic matter (TOM), total bacteria (TB), and SRB. Possible methylation processes of mercury in mangrove sediments were also discussed.

1 Materials and methods

Samples were collected in July and August 2007. Four surface sediment samples and one sediment core (site 4) were taken in the mangrove forest (Fig. 1). Surface sediments were collected using plastic spoons, and the sediment core was taken by a PVC tube with a diameter of 7.5 cm. All sediment samples were stored in sealed plastic bags and kept in an ice box. To avoid Hg contamination, all tools were cleaned beforehand with HNO_3 and rinsed with double distillated water. A fraction of fresh sample was separated to analyze TB and SRB. Samples were air dried and weighed to obtain dry mass. After drying and disaggregation, the sediment samples were ground with an agate mortar and sieved with a 100-mesh sieve.

Samples were digested with concentrated HNO_3, concentrated H_2SO_4, and 5% $KMnO_4$ (Ding and Wang, 2003). F732-V cold vapor atomic absorption spectrometry (CVAAS) (Daji, China) was used to measure THg. Moisture, pH, and TOM were measured according to ISSCAS (1987). TOM was determined using the potassium dichromate oxidation method. Sediment pH values were detected at the ratio of sediment and water 1:2.5. Salinity was measured at the ratio of water and sediments 1:5. Total sulphur (TS) content was determined by Barium sulfate turbidimetry spectrophotometry (UV-1600, Rayleigh, China) (Fu et al., 2007) and sediment MeHg was determined by GC-CVAFS (Glas-Col TM568; Tekran Model 2500, Tekran, USA) with solvent extraction (Liang et al., 1996) following Method 1630 (US EPA, 2001). TB was determined by DAPI dying coupled with UV-fluorescence microscope counting (Fuhrman et al., 1980) and SRB were determined by test bottles as selective medium with MPN counting (SMMOC, 1993). The sediment core was sectioned into depth intervals of 2 cm and then treated the same way as the surface sediments aforementioned.

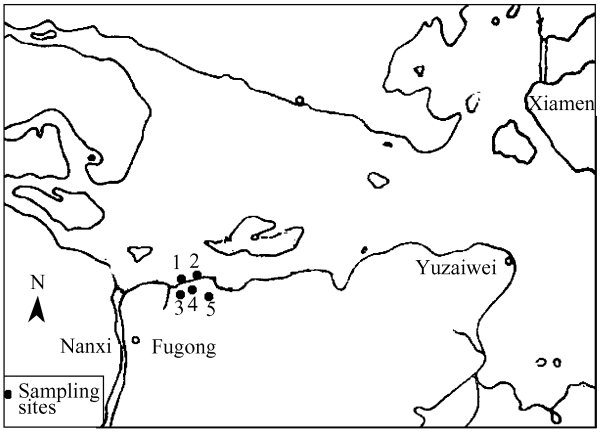


Fig. 1 Study areas and sample sites (1–5).

Quality control for Hg and methyl-Hg determinations was addressed with certified reference materials (GSD-3; IAEA356) and blind duplicates. Limits of determination were 0.01 ng/g for THg and 0.003 ng/g for MeHg in sediment samples. Average THg concentration of the GSD-3 geological standard was 0.016 ng/g (n = 5), which was comparable with the certified value of (0.018 ± 0.002) ng/g. The average methyl-Hg concentration of (5.56 ± 0.54) ng/g (n = 7) was obtained from IAEA356 with the certified value of (5.4 ± 0.89) ng/g. The percentage of recoveries on spiked samples ranged from 95.5% to 118.8% for methyl-Hg in sediment samples. All measurements were calculated on an air-dried basis. All data were analyzed using SPSS 16.0 for Windows.

2 Results

2.1 Distributions of THg, MeHg, and SRB in surficial sediments

The Hg concentrations and environmental parameters of surface sediments are listed in Table 1. The contents of THg and MeHg in surficial sediments were (350 ± 180) ng/g and (0.47 ± 0.11) ng/g (n = 4), respectively, and the MeHg/THg ratio was low, ranging from 0.13% to 0.47%. The amounts of TB and SRB in JRM surface sediments were ($1.4 \times 10^{11} \pm 4.1 \times 10^{9}$) cfu/g dry weight (dw) and ($5.0 \times 10^{6} \pm 2.7 \times 10^{6}$) cfu/g dw ($n$ = 4), respectively, with SRB constituting only a very small fraction of TB. The content of THg in JRM was lower than that determined by Ding et al. (2009), which may be the result of different sampling sites.

Compared with other estuaries in Europe (Table 2), JRM is seriously contaminated with Hg. Significant Hg contam-

Table 1 Content of Hg and environmental factors in surface sediments from Jiulong River Mangrove Provincial Reserve (JRM)

Items	Range	Mean ± SD
SRB ($\times 10^6$ cfu/g dw)	2.5–7.8	5.0 ± 2.7
TB ($\times 10^{11}$ cfu/g dw)	1.0–1.9	1.4 ± 4.1
TOC (%)	2.8–5.1	3.80 ± 0.99
pH	6.6–7.2	6.9 ± 0.3
TS (%)	0.07–0.75	0.32 ± 0.30
Salinity (‰)	5.1–16.5	9.7 ± 5.5
THg (ng/g)	170–620	350 ± 180
MeHg (ng/g)	0.23–0.80	0.47 ± 0.11
MeHg/THg (%)	0.13–0.47	0.26 ± 0.15

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Table 2 Contents of THg and MeHg in sediments from different estuaries

Estuary	Hg (mg/kg)	MeHg (μg/kg)	Reference
Scheldt (Belgium)	0.14–1.80	0.8–6	Baeyens et al., 1998
Ore (Sweden)	0.03–0.12	0.01–1.00	Kwokal et al., 2002
Krka (Croatia)	0.10–1.42	0.01–1.40	Kwokal et al., 2002
Tagus (Portugal)	0.01–66.7	0.3–43	Canario et al., 2005, 2007
British estuaries (UK)	0.05–4.46	0.1–4.0	Craig and Moreton, 1986
Medway (UK)	0.02–1.30	–	Spencer et al., 2006
Lot-Garonne (France)	0.06–0.5	–	Schafer et al., 2006
Adour (France)	0.004–1.46	0.1–1.6	Stoichev et al., 2004
Seine (France)	0.3–1.0	0.1–6.0	Mikac et al., 1999
Medway – Horrid Hill (UK)	0.02–1.2	0.02–4.3	Ouddane et al., 2008
Jiulong River Estuary (China)	0.25–0.59	–	Ding et al., 2009
Jiulong River Estuary (China)	0.17–0.62	0.23–0.80	This study

ination in estuaries such as Scheldt (Belgium) and Tagus (Portugal) resulted from regional industrial development (Baeyens et al., 1998; Canario et al., 2005). With rapid economic growth in China, many factories have been built since the 1990s, from which large amounts of wastewater and solid waste, together with the widely used Hg-bearing pesticides in aquaculture, have been discharged to the mangrove wetlands without treatment (Xue, 2005). These industrial and aquacultural activities may lead to the high load of Hg in mangrove sediments.

2.2 Distributions of THg, MeHg, and SRB in the sediment core

Distributions of THg, MeHg, TB, SRB, and some environmental factors in the sediment core are shown in Fig. 2. Generally, THg, MeHg, TB, salinity, MeHg/THg, and TS increased with depth, but TOM, SRB, and pH decreased.

The contents of THg and MeHg increased irregularly with depth. Average THg concentration was (240 ± 24) ng/g, significantly exceeding the background value of 25 ng/g for sediments of the China shelf sea (Zhao and Yan, 1994). The highest THg level was 290 ng/g at a depth of 6–8 cm, while the lowest was 210 ng/g at 0–2 cm (Fig. 2a). Average MeHg concentration was (0.30 ± 0.15) ng/g, and reached a maximum value of 0.53 ng/g at 14–16 cm (Fig. 2b). MeHg/THg were significantly elevated in the sediment core, and reached a maximum at a depth of 24–26 cm (Fig. 2c).

Based on the average sedimentation rate of 35.42 mm/year in JRM (Tan and Zhang, 1997), we reconstructed Hg pollution history by vertical Hg distribution in the sediment core. Due to much stricter environmental protection policies in recent years, both THg and MeHg concentrations in sediment have decreased in the last decade (Fig. 2a), but they showed significant annual variation.

In the sediment core, TB activity intensities increased with depth (Fig. 2d), but SRB decreased with depth (Fig. 2e). Highest SRB activity intensity reached 7.8×10^6 cfu/g dw at a depth of 2–4 cm, with the second highest value of 3.1×10^6 cfu/g dw at 22–24 cm (Fig. 2a). Both SRB and TB reached their highest values (or second peak value) at a depth of 24–26 cm. This probably results from root cell secretions providing a suitable micro-environment for microbes in the rhizosphere (Clark et al., 1998). Guimaraes et al. (2000) also found that root exudates and decomposing root tissues affected microbiological activities and Hg methylation.

Concentrations of TOM in sediment were relatively high from the surface to a depth of 14 cm but decreased irregularly downwards, with the lowest level of 2.9% found at 24–26 cm (Fig. 2f). The decomposition of mangrove forest litter increases organic detritus at the surface sediments, which leads to an increase in TOM content in the upper-section (Lu and Lin, 1988).

Sediment depth of 24–26 cm corresponded to the peak value of TB, MeHg, MeHg/THg, and the minimum value of TOM. This possibly indicated that mangrove root excretions were bioavailable for microbial activities and created a suitable geochemical environment for Hg methylation. Marvin-DiPasquale et al. (2003) found that sediments located around rooted macrophytes were the most active zones for MeHg production. The Hg methylation in root zones occurs mainly in the root-associated solids and Hg methylation in macrophyte roots is carried out by microorganisms attached to the roots and their diverse associated solids (Guimaraes et al., 2000).

The TS concentrations increased with depth. Sulfur content ranged from 0.01%–0.19%, with an average value of 0.06% and a maximum value of 0.19% at 26–28 cm (Fig. 2g). Ranging from 6.3 to 6.6, the pH value decreased slightly with depth (Fig. 2h). Conversely, salinity increased significantly with depth. It ranged from 7.5‰ to 30‰ and reached its highest value (30‰) at a depth of 26–28 cm and its lowest level (7.5‰) at 2–4 cm (Fig. 2i).

3 Discussion

The methylated form of Hg in estuaries and its subsequent bioaccumulation in edible aquatic organisms represent a major pathway for human exposure to MeHg (Ouddane et al., 2008). Consequently, it is important to determine the factors controlling MeHg production in estuary sediments. Geochemical parameters (e.g., redox, sulfide, pH, and organic content) in sediment are more important than absolute THg concentration for controlling MeHg production (Marvin-DiPasquale et al., 2003). Sulfur, organic carbon, and other sediment compositions can affect MeHg production by changing the amount of bioavailable inorganic Hg and by stimulating methylating microbial activities (Sunderland et al., 2006). Geochemical

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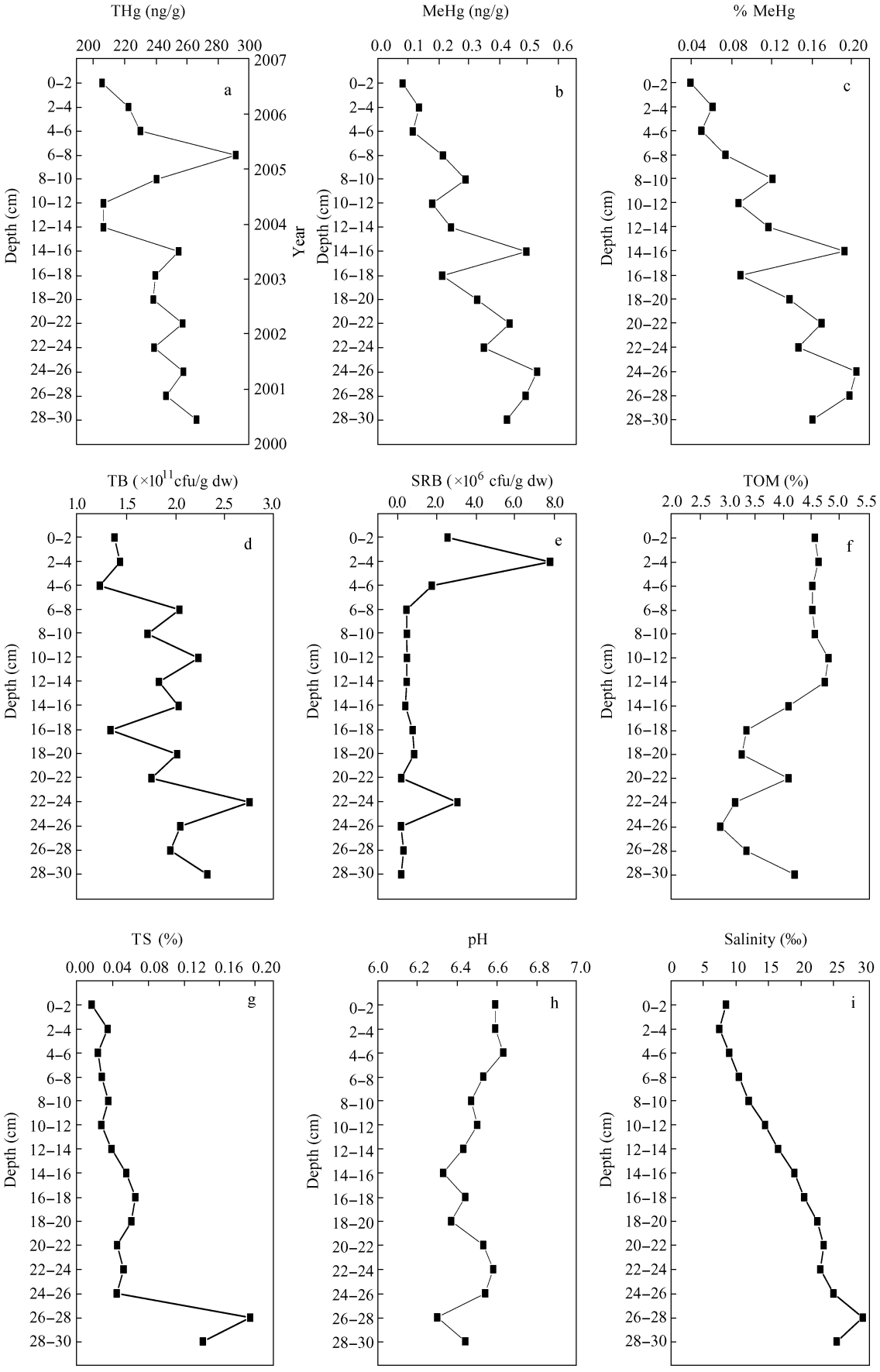


Fig. 2 Distribution of THg, MeHg, and environmental factors in the JRM sediment core.

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Table 3 Correlation matrix of MeHg and environmental factors in the sediment core (n = 15)

	TOM	pH	Salinity	THg	TS	MeHg	SRB	TB
pH	0.281	1						
Salinity	–0.758**	–0.583*	1					
THg	–0.333	–0.183	0.366	1				
TS	–0.414	–0.650**	0.729**	0.303	1			
MeHg	–0.614*	–0.564*	0.852**	0.558*	0.577*	1		
SRB	0.183	0.513	–0.605*	–0.505	–0.397	–0.695**	1	
TB	–0.323	–0.314	0.561*	0.340	0.311	0.588*	–0.374	1
MeHg/THg	–0.614*	–0.598*	0.867**	0.450	0.575*	0.990**	–0.673**	0.600*

** Correlation is significant at the 0.01 level (2-tailed); * correlation is significant at the 0.05 level (2-tailed).

factors controlling Hg methylation can be divided in two groups. The first group impacts Hg bioavailability and the other group impacts the activities of the methylating bacteria (Winfrey and Rudd, 1990; Choi and Bartha, 1993). To understand Hg methylation in mangrove wetland sediments, correlations between MeHg concentrations and environmental factors such as TOM, pH, TS, S, TB, and SRB were discussed. The correlation coefficients are listed in Table 3.

3.1 THg and MeHg/THg

There was a significant positive correlation between THg and MeHg in the sediment core ($r = 0.558$, $p < 0.05$) as shown in Table 3, but the highest MeHg content did not occur at the depth of 6–8 cm where THg reached its peak. This may be due to Hg methylation being affected by high concentrations of oxygen and low activities of methylators in the surface layer. Ouddane et al. (2008) found that stable anoxic conditions are generally more favorable for Hg methylation than oxic conditions. Hammerschmidt and Fitzgerald (2006) also found that surface sediment MeHg is positively correlated to inorganic Hg (Hg(II) = total Hg – MeHg) concentrations.

MeHg/THg represents the fraction of THg potentially available for conversion to MeHg, and thus reflects the degree of Hg methylation (Sunderland et al., 2006). Results showed that sediment core MeHg/THg ranged from 0.04% to 0.21%, and was relatively low in the upper section (0–8 cm) and relatively high in the lower section (Fig. 2c). This suggests that a high degree of methylation occurred at the bottom, which is in agreement with previous studies (Ouddane et al., 2008). In addition, higher proportions of MeHg did not register in the most contaminated sediments, presumably reflecting low methylation rates and/or favorable demethylation reactions near the sediment-water interface as well as volatilization processes (Bubb et al., 1993). Both MeHg concentrations and MeHg/THg were significantly elevated in the sediment core.

3.2 TOM and TS

Generally, organic matter can stimulate microbial activities, reduce oxygen levels, decrease Hg(II) bioavailability, and therefore increase biomethylation (Barkay et al., 2003). Research has shown that humus leads to Hg methylation in freshwater lakes (Weber, 1993), Hg methylation ratio and TOM content show significant positive correlation (Ullrich et al., 2001), and organic material largely controls spatial distributions of Hg(II) and MeHg (Hammerschmidt et al., 2008).

In the present study, TOM content showed a significant negative correlation with MeHg in the sediment core (r = –0.614, $p < 0.05$) (Table 3). This indicates that TOM in mangrove sediments cannot be utilized completely by SRB and cannot excite activities of Hg methylators. This might be due to the effects of high salinity on the bioavailability of TOM and the activities of functional groups, particularly as allochthonous organic material (terrestrial and/or sewage) and dissolved sulfide reduce bioavailability of Hg and attenuate MeHg production in sediment (Hammerschmidt et al., 2008).

The content of TS in JRM was generally high, and showed significant positive correlation with MeHg ($r = 0.577$, $p < 0.05$) (Table 3), suggesting that sulfur content could accelerate net Hg methylation. Higher sulfide concentrations appear to yield a geochemical environment conducive to Hg(II) uptake by methylating bacteria and correspond to an elevated fraction of Hg in methylated form (Sunderland et al., 2006). High concentrations of sulfide and dissolved organic matter can provide a favorable environment for microbial methylators to absorb easily (Sunderland et al., 2006).

3.3 pH

There was a significant negative correlation between MeHg concentration and pH ($r = –0.564$, $p < 0.05$) (Table 3). The pH values in sediment had an important effect on solubility, redox, precipitation, dissolution, and adsorption of Hg, and therefore impacted Hg speciation. Previous studies have shown that the most suitable pH value for Hg methylation was 5.0 (Chen et al., 2005). The pH in this study area ranged from 6.3 to 6.7 and decreased with depth, which is important as even small pH (7.3–6.3) changes can result in large increases in Hg(II) uptake by bacteria (Kelly et al., 2003). The increased rate of bioaccumulation was directly proportional to the concentration of H^+.

3.4 Salinity

Salinity showed a significant positive correlation with MeHg concentrations ($r = 0.852$, $p < 0.01$) (Table 3). In the sediment core, high MeHg content always corresponded to high salinity and high sulfide concentrations. High MeHg/THg was observed even though salinity reached 30‰. Ouddane et al. (2008) found that methylation was favored in estuaries where SO_4^{2-} could not act as a restraining factor due to high salinity compared to

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freshwater. While previous studies have mainly focused on Hg methylation in lakes, reservoirs, and other freshwater wetlands, little is known about Hg methylation in mangrove wetlands with relatively high salinity. The effect of salinity on Hg methylation cannot be neglected. Considering the large difference between freshwater and estuary wetlands, it is expected that different Hg methylation mechanisms exist for different wetlands.

3.5 TB and SRB

Previous studies indicated that SRB were the main Hg methylators in freshwater sediments and anaerobic estuary sediments (Compeau and Bartha 1985; Choi and Bartha, 1993; King et al., 1999, 2000), and factors that affect SRB activity influence Hg methylation (Choi and Bartha, 1993). Therefore, methylation potential in sediments depends on substrate availability and SRB activity (Gilmour and Henry, 1991).

Although only one sediment core was sampled, data obtained demonstrated significant negative correlation between SRB and MeHg. No significant correlations between SRB and MeHg were found from surface sediments in the eight mangrove areas of China (Ding et al., 2010). This suggests that Hg methylation in mangrove sediments is unique.

Contrary to previous studies, SRB may be not the main Hg methylators as other micro-organisms may contribute to Hg methylation in the mangrove sediments of this study. The reasons for this are: (1) SRB only constituted a very small fraction in TB, and TB contents were 5–6 orders higher in magnitude than those of SRB; (2) there was a significant negative correlation between MeHg content and total number of SRB ($r = –0.695$, $p < 0.01$); and (3) there was a significant positive correlation between MeHg content and total number of TB ($r = 0.588$, $p < 0.05$) (Table 3).

Many convincing arguments have shown that SRB, responsible for MeHg production, may not be relevant to marine waters (Weber, 1993). The high sulfur concentrations in estuaries and seawater cause Hg to bond to reductive sulfur (S^{2-}), which makes it unavailable for methylation (Hammerschmidt et al., 2008; Han et al., 2008). An investigation on the influence of sulfide concentration on biotic methylation of Hg(II) confirmed, however, there is an inverse correlation between sulfide concentrations and biotic MeHg production (Benoit et al., 1999). It is difficult, therefore, to explain the widespread occurrence of MeHg in marine biota by methylation through SRB. It is interesting to note that SRB can methylate Hg and demethylate Hg at different rates simultaneously (King et al., 1999; Duran et al., 2008).

Saprophytic fungi have been found to methylate Hg in terrestrial environments (Fischer et al., 1995). The redox potential decreases with depth compared to SRB, however, and other extreme anaerobic microbes such as methanogens bacteria and acetate reducing bacteria can only survive in more reduced conditions and thus show high Hg methylation capacity in deep sediment (Stumm and Morgan, 1996). Ramamoorthy et al. (1982) also discovered that dead bacteria promoted Hg methylation through emission of enzymes to water. Parkman et al. (1995) found that extracellular enzymes could accelerate Hg methylation. The increased number of TBs might have an underlying promotional effect on Hg methylation. Conversely, other research has found that demethylation of MeHg occurred in nature and/or under the effects of some microbes (Parkman et al., 1995; Guimaraes et al., 2000). Many bacteria predominated by aerobic microbes, including living and dead, can lead to demethylation (Oremland et al., 1991).

4 Conclusions

(1) The contents of THg, MeHg, TB, and SRB in surficial sediments were (350 ± 150) ng/g, (0.47 ± 0.11) ng/g, $(1.4 \times 10^{11} \pm 4.1 \times 10^{9})$ cfu/g dw and $(5.0 \times 10^{6} \pm 2.7 \times 10^{6})$ cfu/g dw, respectively. Industrial and aquacultural activities are the main reasons for Hg pollution in the mangrove wetland. (2) The contents of THg, MeHg, TB, and SRB in sediment core were (240 ± 24) ng/g, (0.301 ± 0.148) ng/g, $(1.9 \times 10^{11} \pm 4.2 \times 10^{10})$ cfu/g dw and $(1.3 \times 10^{6} \pm 2.0 \times 10^{6})$ cfu/g dw, respectively. THg, MeHg, TB, MeHg/THg, salinity, and TS increased with depth, and TOM, SRB and pH decreased with depth. (3) MeHg concentrations in sediments were significantly positively correlated with THg, salinity, TS, and MeHg/THg, and significantly negatively correlated with SRB, TOM, and pH.

Because only one sediment core was sampled in this study, future research is needed to verify contributions of sulfur speciation, dissolved organic matter content, and possible Hg methylators in JRM.

Acknowledgments

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