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ORIGINAL PAPER

Distribution and assessment of mercury (Hg) in surface sediments of Futian mangrove forest, China

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Abstract To investigate the distribution of mercury (Hg) in Futian mangrove wetland, surface sediments from land to sea were collected, including *Kandelia obovata*, *Avicennia marina*, *Sonneratia caseolaris*, and mud flat. The ecological risks of Hg in sediments were also assessed. The results showed that mangrove forests acidified sediments and promoted the accumulation of salinity and organic matter in sediments. Hg concentrations in both mangrove forests (154.7–218.4 ng g^{-1}) and mud flat sediments (226.3–251.9 ng g^{-1}) surpassed the background level (71.0 ng g^{-1}). Furthermore, Hg concentrations in sediments decreased gradually from sea to land at all depth. From the bottom to the top layer sediment, Hg concentration decreased gradually in the sediments near land, while it kept vertically stable in the coastal area, indicating its pollution may mainly come from the coastal area rather than the land to some extent. Although the mean values of geo-accumulation indexes revealed uncontaminated to moderately contaminated levels, the mean values of potential ecological risk coefficients revealed considerable ecological risk of Hg to the environment, deserving further attention.

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Keywords Mercury · Distribution · Futian mangrove sediment · Ecological risk assessment

Introduction

Mangrove forests thrive widely in the intertidal zones on most tropical and subtropical coasts (Conrad et al. 2017). As one of the most productive ecosystems in the world, mangroves not only provide wood, food, and medicines for human beings (Walters et al. 2008), but also stabilize fine sediments, prevent coastal erosion, and maintain water quality and biodiversity (Arrivabene et al. 2016). Moreover, the mangrove sediments have a large capacity to retain heavy metals and reduce the potentially toxicological effects due to the characteristics of fine-grained texture, low redox condition as well as enrich of sulfide and organic matters (Chai et al. 2015; Gordeeva et al. 2017). Mercury (Hg) is widely considered to be one of the most toxic heavy metals due to its strong biological toxicity and complex migration process among ecosphere, atmosphere, hydrosphere, and biosphere (Luo et al. 2012; Jardine et al. 2013; Li et al. 2016a). It has been reported that Hg has no known essential biological function (Chakraborty et al. 2015), but elevated Hg levels in human bodies can lead to serious damage on health, resulting in neurological, immunological, and nephrological disorders, etc. (Cornelis et al. 2005; Zahir et al. 2005; Dhivert et al. 2016). At present, more

and more researches are conducted to investigate the distribution, speciation, and transportation of Hg in mangrove sediments, where surroundings were heavily affected by human activities (Kehrig et al. 2012; Machado et al. 2016; Haris et al. 2017).

Futian mangrove forest in Shenzhen Bay is the only mangrove located within the large cities of China, which is deeply influenced by human activities. Many studies found that with the rapid urbanization and industrialization of Hong Kong, Shenzhen, and other coastal cities of Pearl River Estuary (PRE) since the early 1990s, a large amount of industrial wastewater has been discharged into the Shenzhen Bay (Huang et al. 2003; Zuo et al. 2008; Li et al. 2012; Chan et al. 2016), and abundant heavy metals have been deposited in Futian mangrove sediments simultaneously (Tam and Wong. 2000; Zan et al. 2002; Tang et al. 2016). Nowadays, some studies have investigated Hg and methyl Hg, as well as Hg flux in Futian mangrove forest sediments with low sampling density (Ding et al. 2009, 2010a; Li et al. 2016a). In the sediment of Futian mangrove forest, the spatial distributions of heavy metals (Cu, Cr, Cd, Ni, and As) varied in different mangrove communities from land to sea (Li et al. 2016b). Therefore, the distance from land and the type of mangrove communities may also act as an important factor in affecting Hg accumulation in sediments (Bayen 2012). However, no systematic and specialized research is available on the distribution and assessment of Hg in surface sediments of Futian mangrove forest with high sedimentation rate (Li et al. 2012). The data are also basic in making the long-term management and conservation policies. Therefore, the objectives of this study were to (1) determine the distribution of Hg and physicochemical properties in surface sediments of different mangrove communities and mud flat from land to sea and (2) assess the current potential ecological risk of Hg.

Materials and methods

Study area

The study was carried out in the Futian Mangrove National Natural Reserve (22°32′N, 114°03′E, Fig. 1), located in the northeast coast of Shenzhen Bay, Guangdong province, China. The Futian Mangrove National Reserve covers an area of 367.64 ha distributed along 9 km of the coast. The mean annual temperature is 22 °C, the mean annual precipitation is 1927 mm mostly between May and September, and the tides in Shenzhen Bay are semidiurnal, with an average range of 1.9 m. There are three rivers flow into the mangrove reserve directly, including Shenzhen river, Xinzhou river, and Fengtang river (Zan et al. 2002). The mangrove forest communities distribute with a 100–400-m width from land to sea, including *Kandelia obovata*, *Avicennia marina*, *Sonneratia caseolaris*, and mud flat, respectively (Xie et al. 2010).

Sample collection and sediment analysis

According to sedimentation rate (1.38 cm a^{-1}) in Futian mangrove forest (Li et al. 2012), the top 0–20 cm depth of sediments would encompass the deposited sediments after 2000s. In December 2015, three mangrove communities and mud flat were chosen in Futian mangrove forest, China. In each of these four swamps, three sampling sites were distributed along the coastline. In each sampling site, three surface sediments (0–20 cm depth) were collected with a 10-cm-diameter PVC tube which was washed by deionized water after soaking by dilute nitric acid. Then, each sediment core was sliced into 5-cm layers. Three subsamples at the same depth interval were collected to mix a composite sample in a plastic tray. Thus, there are 48 sediment samples in total (Fig. 1). All sediment samples were immediately sealed with plastic bags and transported back to the laboratory on the same day.

The sediment samples were air-dried, powdered, and sieved through 0.25 mm sieve. The sediment pH and salinity were determined in deionized water using mass ratios of 1:5 (sediment to water) (Li et al. 2016a). The total organic carbon (TOC) was determined by multi N/C 3100 analyzer (Jena, Germany). The particle size was analyzed by a particle size analyzer (Mastersizer 3000, Malvern, UK). The bulk sediment samples were separated into fine particles (clay + silt) (< 63 μm) and coarse particles (> 63 μm) (Parthasarathi et al. 2015). To determine Hg concentrations in sediment, 0.1 g of sediment samples was subjected to Milestone ETHO microwave digestion in a mixture of 9 ml nitric acid (HNO_3), 3 ml hydrofluoric acid (HF), and 1 ml hydrochloric acid (HCl). The Hg concentration in extracting solution was analyzed by using

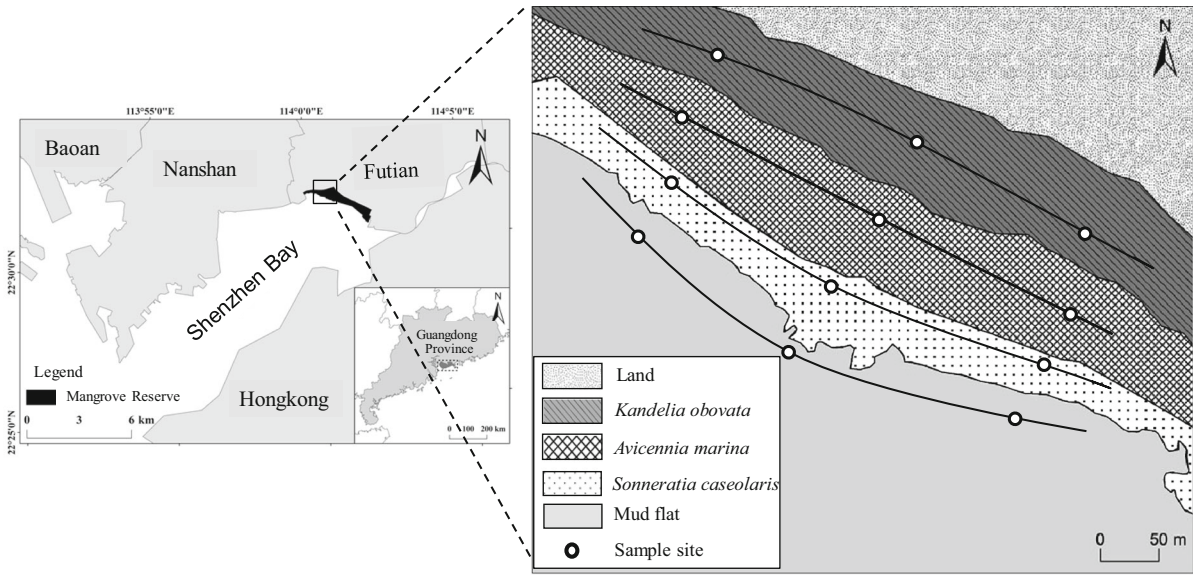

Fig. 1 Sampling sites in Futian mangrove forests of Shenzhen Bay, China

manual atomic fluorescence spectrophotometer (Brooks Rand Labs-Model III, America).

Ecological risk analysis

The geo-accumulation index (I_{geo}) was implied to calculate the enrichment of Hg. The I_{geo} was introduced by Müller and can effectively explain the heavy metal pollution level by anthropogenic activities (Müller 1969) The I_{geo} is defined as: $I_{geo} = \log_2(C_n/1.5B_n)$, where C_n is Hg concentration in sediments and B_n is the background concentration of Hg. The constant factor 1.5 is the background matrix correction factor due to lithogenic effects. The I_{geo} result can be interpreted into seven classes (Table 1).

The potential ecological risk coefficient (E_r^i) was chosen to evaluate the ecological risk of Hg. The method was proposed by Hakanson (1980), and it combined the content and the biological toxicity comprehensively. The E_r^i of Hg can be defined as: $E_r^i = C_n/B_n{*}T_n$, where C_n is Hg concentration in sediments; B_n is the background concentration of Hg; and the T_n is the biological toxicological coefficients of Hg ($T_n = 40$). The degree of ecological risk can be categorized as follows: $E_r^i < 40$, low risk; $40 \le E_r^i < 80$, moderate risk; $80 \le E_r^i < 160$, considerable risk; $160 \le E_r^i < 320$, high risk; $E_r^i \ge 320$, very high risk.

Statistical analysis

The data were expressed as mean ± standard deviation. Data statistics and analysis were conducted with Excel 2013. The Spearman correlation analysis and

Table 1 Pollution level classification of the index of geo-accumulation (I_{geo})

Value	Class	Pollution quality
$I_{geo} \le 0$	0	Uncontaminated
$0 < I_{geo} < 1$	1	Uncontaminated to moderately contaminated
$1 < I_{geo} < 2$	2	Moderately contaminated
$2 < I_{geo} < 3$	3	Moderately to heavily contaminated
$3 < I_{geo} < 4$	4	Heavily contaminated
$4 < I_{geo} < 5$	5	Heavily to extremely contaminated
$I_{geo} \ge 6$	6	Extremely contaminated

two-way ANOVA were performed at 5% level by SPSS 16.0 statistics package (SPSS Inc., Chicago, IL, USA).

Results and discussion

Sediments characteristics

Sediment, the primary compound of mangrove ecosystem, could accumulate various contaminants from incoming tidal waters and freshwater sources (Marchand et al. 2016; Zhang et al. 2018). The physicochemical properties of the sediments have been proved to be important in affecting Hg accumulation, formation, and transportation (Machado et al. 2002; Shi et al. 2010; Zhou et al. 2010). In Fig. 2, the pH in sediments varied from 5.8 to 6.6 in mangrove forest, slightly lower than that in mud flat (6.5–6.8). These results are consistent with the previous studies (Zhou et al.2010; Li et al. 2016c), suggesting that mangrove forests could acidify sediment due to microbial decomposition of mangrove litter and oxidation of FeS_2 and FeS (Tam and Wong. 2000). The salinity in all sediments ranged from 3.7 to 6.2‰, and the salinity in *K. obovata* and *A. marina* sediments was much higher than those in mud flat, which may be related to their strong ability of salt accumulation (Wang and Lin 2003). TOC content in sediments significantly decreased from land to sea. TOC content in the sediments of *S. caseolaris* and *A. marina* slightly increased in the surface layer, which may be derived from the decomposition of litter and the metabolism of algae on the surface sediments (Zhou et al. 2010; Li et al. 2016a; Machado et al. 2016). In particular, TOC content (5.2–6.3%) in *K. obovata* sediment is significantly higher than most of the mangrove wetlands in China (1.2–3.8%) (Ding et al. 2010b). The large fluctuation of salinity and the high TOC content in *K. obovata* sediments may relate to that *K. obovata* is close to land and significantly affected by the input of domestic sewage (mainly from Fengtang river) and runoff from land (Zhang et al. 2000; Liu et al. 2014). The sediments in the study area mainly consisted of fine particles (< 63 um), with strong heterogeneity, and no significant changes from land to sea have been observed, indicating the similar hydrologic condition and depositional environment (Zan et al. 2002). It is noteworthy that the content of fine particles in the *S. caseolaris* sediments gradually decreased from the bottom to the top layers, which was possibly related to the baffle effect of roots and trunks. The high density respiration roots could effectively reduce the hydrologic movement of tides (Liu et al. 2008), which facilitate the deposition of larger particulates in tides. The result of two-way ANOVA showed that sediment pH and salinity were significantly affected by mangrove community and sediment depth; sediment TOC content was significantly affected by mangrove community and its interaction with sediment depth ($P < 0.001$) (Table 2).

Hg accumulation in surface sediments

In Table 3, Hg concentrations in mangrove forests and mud flat sediments were 154.7–218.4 and 226.3–251.9 ng g^{-1}, respectively, which were higher than the background value of 71 ng g^{-1} (Ding et al. 2009). The higher Hg concentration was consistent with previous studies (Ding et al. 2009; He et al. 2015; Li et al. 2016a), which may be related to the input of industrial wastewater from neighboring regions since 2000s. And, it was higher than many mangrove wetlands, such as Shankou, Beilun estuary, and Yunxiao in China (Ding et al. 2009), as well as Port Klang in Malaysia (Haris et al. 2017) and Sundarban in India (Kwokal et al. 2012). Furthermore, Hg concentration in this study was lower than Fugong, Quanzhou bay (Ding et al. 2009) in China and Guanabara in Brazil where the environments were strongly influenced by anthropogenic activities (Machado et al. 2002). Compared with the guideline values, Hg concentrations were close to the threshold effect concentration (TEC), but much lower than the probable effect concentration (PEC), indicating low threat of Hg to the organisms in mangrove ecosystem.

Hg concentrations in mud flat were higher than that in mangrove communities and decreased gradually from sea to land at all the same depths (mud flat > *S. caseolaris* > *A. marina* > *K. obovata*) (Table 3, Fig. 3). From the bottom to the top layers of sediment, Hg concentration in sediments decreased gradually in *K. obovata* and *A. marina* sediments which are close to the land, while stable trends of Hg concentrations were detected in *S. caseolaris* and mud flat sediments near the coastal area (Fig. 3). Similar conclusions have also been reported by Li et al. (2015) that the concentrations of Cu, Pb, and Zn in surface sediments

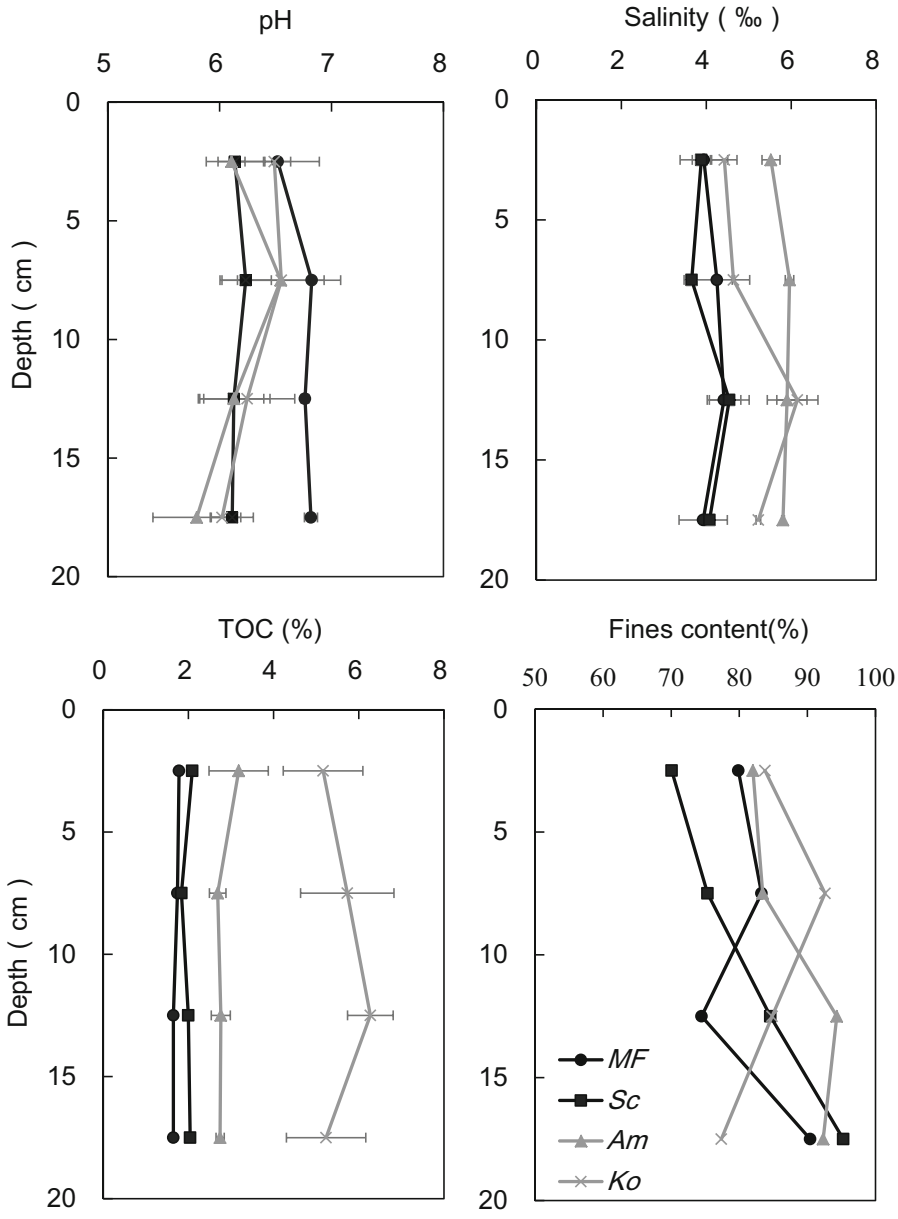

Fig. 2 Vertical profiles of physicochemical properties in sediments from the *K. obovata* (*Ko*), *A. marina* (*Am*) *S. caseolaris* (*Sc*), and mud flat (*MF*) sites

Table 2 Two-way ANOVA of effect of mangrove community, sediment depth, and their interactions on Hg accumulation and physicochemical properties in sediment

	Hg	TOC	pH	Salinity
Mangrove community	8.5***	332.0***	20.9***	20.4***
Sediment depth	0.3	1.2	5.9**	3.5**
Mangrove community × Sediment depth	0.8	2.5**	2.1	0.8

Data represent F values at 0.05 level. *$P < 0.05$, **$P < 0.01$, ***$P < 0.001$

Table 3 Hg concentrations in Futian mangrove sediments and in other related studies in and out of China

Location	Hg (ng g^{-1} dw)	References
Kandelia obovata	154.7–192.6 (175.6)[a]	This study
Avicennia marina	166.4–211.4 (191.5)	This study
Sonneratia caseolaris	195.7–218.4 (210.6)	This study
Mud flat	226.3–251.9 (237.1)	This study
Shenzhen, China	172.3–192.9 (179.9)	Ding et al. (2009)
Shenzhen, China	159.8–226.7 (175.8)	He et al. (2015)
Shenzhen, China	92.0–196.0	Li et al. (2016a)
Shankou, China	51.6–107.9 (73.8)	Ding et al. (2009)
Beilun estuary, China	102.5–217.6 (117.8)	Ding et al. (2009)
Yunxiao, China	31.3–168.6 (63.3)	Ding et al. (2009)
Fugong, China	248.1–591.4 (438.2)	Ding et al. (2009)
Quanzhou bay, China	409.6–543.6 (467.5)	Ding et al. (2009)
Guanabara, Brazil	260.0–890.0	Machado et al. (2002)
Port Klang, Malaysia	0.2–150.0 (28.5)	Haris et al. (2017)
Sundarban, India	7.3–93.3 (26.1)	Kwokal et al. (2012)
Background value, China	71.0	Ding et al. (2009)
Threshold effect concentration (TEC)	180.0	Vane et al. (2009)
Probable effect concentration (PEC)	1060.0	Vane et al. (2009)

The background value, TEC, and PEC of mercury were also shown for comparison

TEC, threshold effect concentration, indicates concentrations below which adverse effects on biota are rarely observed. *PEC,* probable effects concentration, indicates concentrations above which adverse effects on biota are frequently observed

[a]Concentration of Hg: min–max (mean)

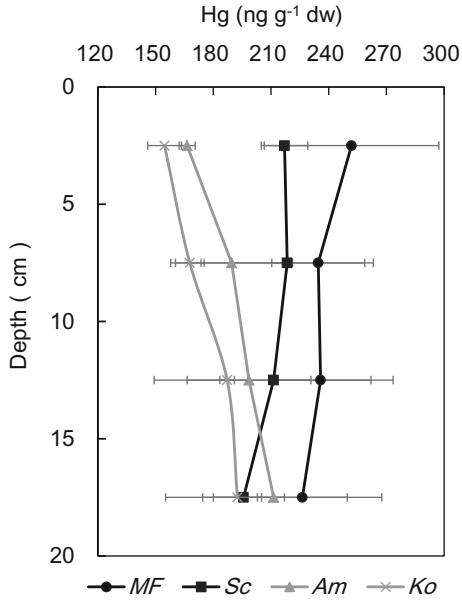

Fig. 3 Vertical profiles of Hg in sediments from the *K. obovata* (*Ko*), *A. marina* (*Am*), *S. caseolaris* (*Sc*), and mud flat (*MF*) sites

(10–20 cm) were slightly higher than the surface sediments (0–10 cm) in Futian mangrove forest near the land. One possible explanation may be that a series of strict environmental protection measures have been carried out, including introducing sewage treatment facilities and relocation of polluting industries from Shenzhen since 2000s (Li et al. 2016c). In addition, the results of two-way ANOVA indicated that the concentrations of Hg were significantly affected by different mangrove communities ($P < 0.001$) (Table 2).

With higher organic matter and fine particulates content, sediment in mangrove forest could retain more heavy metals compared to mud flat (Zhou et al. 2010; Liu et al. 2010). However, mangrove plants have relatively low uptake of Hg, with most Hg deposited in sediments (Machado et al. 2016). In Table 4, Hg concentration in sediments was negatively related to TOC ($r = -0.611$, $P < 0.01$), indicating that the sources for Hg and organic matter may be different. In particular, the higher Hg concentration in mud flat sediments indicated its pollution sources coming from marine coastal environment. Although the terrigenous industrial contaminants have been alleviated due to the implementation of environmental protection policies after 2000s (Machado et al. 2002), the marine Hg pollution mainly from the PRE was not controlled and managed effectively. Since 1980s, huge amounts of heavy metal contaminants have been discharged into the PRE, such as 8293 t heavy metal input coming from direct fluvial transport by the Pearl River in 2003 (Li et al. 2007). The

Table 4 Spearman matrix of Hg and physicochemical properties in sediment

	Hg	TOC	pH	Salinity	Fine particles (< 63 μm)
Hg	1.0				
TOC	− 0.6**	1.0			
pH	0.0	− 0.4*	1.0		
Salinity	− 0.3	0.6**	− 0.3	1.0	
Fine particles(< 63 μm)	− 0.4	0.3	− 0.3	0.4	1.0

*Correlation is significant at 0.05 level; **correlation is significant at 0.01 level, $n = 48$

Shenzhen Bay received tremendous amount of PRE water containing abundant fine particles, which is a major carrier for transporting trace metals from the upstream source area to the coastal zone in Pearl River Mouth Basin (Ip et al. 2005; Ye et al. 2012; Zhao et al. 2017). Moreover, Shenzhen Bay is a semi-closed bay with poor hydrodynamic conditions, especially in the mangrove areas near the shore (Tang et al. 2016), resulting in the absorption of Hg by the fine particles and gradual deposition from sea to land. In addition, the correlation among Hg concentration and fine particles was not significant (Table 4), in agreement with previous studies (Bravo et al. 2011; Haris et al. 2017), indicating that fine particles were not an important factor in affecting Hg accumulation in sediment.

Ecological risk of mercury pollution in sediment

To determine the effects of anthropogenic activities on Hg accumulation in sediment, I_{geo} was applied to facilitate the interpretation of sediment quality. Taking the strong biotoxicity of Hg into account, E_r^i was also used to evaluate the possible biological effects of Hg. In the present study, the background concentration of Hg (71.0 ng g^{-1}) was referenced to the study by Ding et al. (2009). Results showed that the distribution of I_{geo} and E_r^i of Hg in sediments was similar to the corresponding concentrations (Fig. 4), with gradually

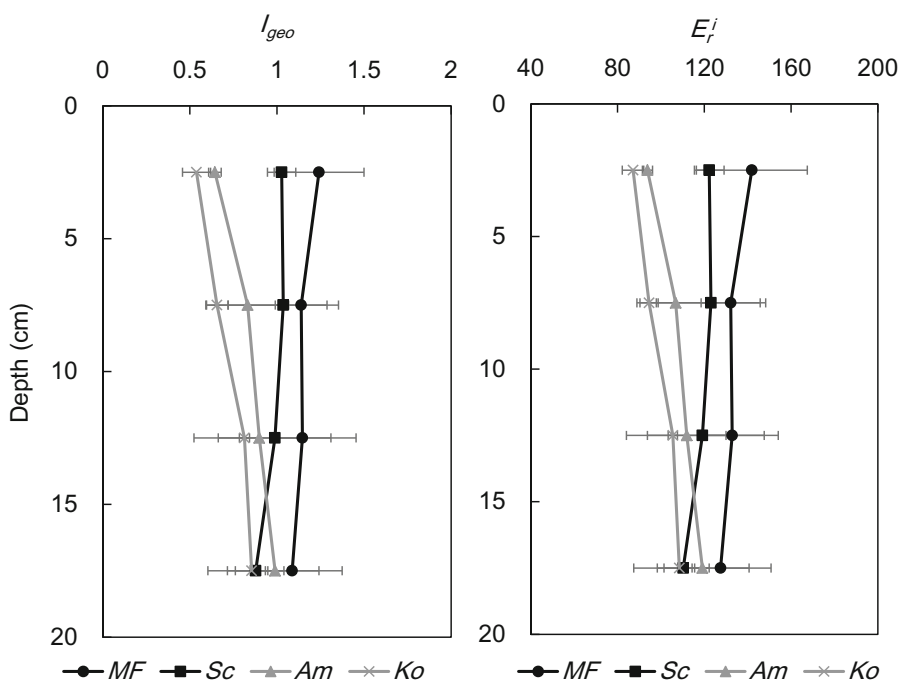

Fig. 4 Vertical profiles of the geo-accumulation index (I_{geo}) and the potential ecological risk coefficient (E_r^i) in sediments from the *K. obovata* (*Ko*), *A. marina* (*Am*), *S. caseolaris* (*Sc*), and mud flat (*MF*) sites

decreased trends from sea to land. Furthermore, reduced trend was also detected from the bottom to the top layers in the sediments of *K. obovata* and *A. marina* marshes. These results may be related to that the anthropogenic metal emission from land has declined as a result of strict environmental policies like improving wastewater treatment facilities and moving polluting industries in the surrounding areas away. The values of I_{geo} in all sediments changed from 0.5 to 1.2 with a mean of 0.9, meaning that the pollution level caused by Hg in sediments ranged from uncontaminated to moderately contaminated. Furthermore, the values of E_r^i varied between 87.1 and 141.9 with a mean of 114.8, suggesting that mercury posed considerable ecological risk to the environment (Fig. 4). Compared with E_r^i values of other heavy metals in Futian mangrove sediments, the E_r^i value of Hg is lower than Cd ($E_r^i \geq 320$), but much higher than that of Cu, Pb, Zn, and Cr ($E_r^i \leq 40$) (Li et al. 2015), which might be the cause for concern.

Conclusions

In this study, sediment in *K. obovata*, *A. marina*, *S. caseolaris*, and mud flat (from land to sea) was analyzed for distribution and ecological risk of Hg in surface sediments of Futian mangrove forest, China. Mangrove forest reduced pH and promoted salinity and TOC content in sediment, compared to mud flat. Totally, Hg concentration in all sediments ranged from 154.7 to 251.9 ng g^{-1}. In the horizontal direction, Hg concentrations reduced from sea to land (from mud flat to *K. obovata*). In the vertical direction, Hg concentration was stable at sea side (mudflat) and decreased at land side (*K. obovata*) from the bottom to the top layer of sediment. Thus, Hg pollutants may mainly come from the marine coastal environment rather than land. The geo-accumulation index (I_{geo}) and potential ecological risk coefficient (E_r^i) revealed that Futian mangrove sediments were polluted by Hg to some extent. Further study should be conducted on the speciation of inorganic Hg and methyl Hg and the bioavailability and toxicity of Hg to organisms.

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