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Distribution and risk of mercury in the sediments of mangroves along South China Coast

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Abstract

The importance of mangrove was widely reported. However, the potential risks of pollutants (e.g., Hg) accumulated in the mangroves are often ignored. Thus, the present study aimed to explore the distribution and risk of mercury (Hg) in the sediments of mangroves along South China Coast. Results showed that concentrations of total Hg ranged from 0.0815 to 0.6377 mg/kg, with an arithmetic mean value of 0.2503 mg/kg. The contamination index (P_i) showed mild pollution toxicity risks in NS, slight toxicity risks in DZG, QZ, SY, ND, GQ, TLG, and free pollutions in BMW, SJ, ZJK and BLHK. NS, DZG and SY scored the highest values of I_{geo} among the eleven mangrove regions studied, indicating moderate to heavy pollution inputs in these regions. As for the distribution of Hg in the sediments along tidal gradient, concentrations of Hg in the sediments sharply increased from seaward mudflat to landward mangrove, corresponding with the increases of TOC. In summary, the present data indicated that mangrove ecosystem is efficient in Hg reservoir. However, the potential ecological risks of Hg, especially in some mangrove regions easily affected by human activities, should be noted.

Keywords Mercury · Mangroves · Pollution assessment · Intertidal gradient

Introduction

Mercury (Hg) is considered as one of the precedence-controlled pollutants due to its high toxicity (Haris et al. 2017; Jia et al. 2017). Hg can directly result in irreversible disease and serious damage in bodies (Ren et al. 2018), such as minamata disease, renal failure as well as nervous system disorder. Although Hg is generally not with high level in natural environment (Jagtap and Maher 2015), Hg has received increasing attention recently and are often found with high concentrations in farmland, rivers, lakes and estuaries due to the rapid development of urbanization and industrialization (Garcia-Ordiales et al. 2017; Men et al. 2018).

Mangroves are one of the most important intertidal estuarine ecosystems along the coastlines in tropical and sub-tropical regions (Luan et al. 2006; Correia and Guimarães 2017; Islam et al. 2017), and have as high productivity, high restitution rate, high decomposition rate and high resistance in the world (Wang 2019). Significant ecological functions of mangroves, such as primary producer, coastal guard and bird habitat, were widely reported (Zhang et al. 2014; Machado et al. 2016). Previous studies have also indicated that mangrove ecosystem is an efficient purifier, since it can receive amounts of contaminants through particle sedimentation during tidal cycle (Tam and Wong 2000). However, the potential risks of pollutants accumulated in the mangroves are often ignored. It should be noted that the accumulation of Hg in mangrove ecosystem may also result in potential exposure risks through biotransformation and biological

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amplification (Guo et al. 2016; Carravieri et al. 2018; Long et al. 2018).

Mangrove sediments are crucial repositories for heavy metals due to rapid sedimentation, besides the chelation between organic matter and contaminants could accelerate the process of pollution fixation and burial (Harbison 1986; Wang et al. 2010; Jiang et al. 2018). Hg pollution in the sediments of mangroves were also widely reported (Ding et al. 2009; Qiu et al. 2011; Wu et al. 2011; Liang et al. 2013; Sun et al. 2017). The status of Hg pollution in mangroves may be varied significantly among different regions. Besides, mangroves often exhibit a special sequential community succession in the intertidal regions (Wang et al. 2019). Generally, relative higher organic matter was often observed in the landward stable mangrove community when compared to seaward pioneer mangroves (Tam and Wong 2000). It was hypothesized that total concentrations of Hg in sediments may also varied associating with the changes of mangrove zonation along tidal gradient.

The southeast coast, the main area of mangrove distribution in China, has witnessed the rapid development of China. As an important buffer between sea and land, mangroves are sensitive and affected by human activities and are subjected to multiple pollution sources (e.g., municipal wastes, framing and mariculture) (Analuddin et al. 2017). Unfortunately, comprehensive assessments of the status of Hg pollution in mangroves with a large spatial scale are still limited. Thus, in this study, eleven mangrove regions were selected to evaluate the status of Hg pollution in mangroves along South China Coast. In terms of the variation of Hg in different tidal positions, four typical mangrove reserves with complete succession zonation were also selected to explore spatial distribution of Hg along tidal gradient.

Therefore, the purposes of the study were (1) to identify the concentrations of Hg in the sediments of eleven mangrove regions along South China Coast; (2) to evaluate the risks of Hg pollutions in eleven mangrove regions along South China Coast; (3) to explore the distribution of Hg and its relations with soil properties along tidal gradient.

Materials and methods

Concentrations of Hg in the sediments of mangroves along South China Coast

During October 2016, eleven regions were selected for the evaluation of mercury pollution in mangroves along South China Coast (Fig. 1): (1) Dongzhai harbor mangrove nature reserve, Hainan province (DZG); (2) Bamen Bay mangrove wetland, Hainan Province (BMW); (3) Tielu harbor, Hainan province (TLG); (4) Sanya, Hainan province (SY); (5) Gaoqiao mangrove reserve, Zhanjiang, Guangdong province (GQ); (6) Beilun estuary, Guangxi province (BLHK); (7) Shijiao mangrove reserve, Guangxi province (SJ); (8) Zhangjiang estuary mangrove reserve, Fujian province (ZJK); (9) Quanzhou, Fujian province (QZ); (10) Ningde, Fujian Province (ND); (11) Nansha mangrove wetland, Guangzhou, Guangdong province (NS). Except ND and TLG, there were ten sampling sites (~30 m from the edge of landward mangrove) at each mangrove region. The areas of mangrove in ND and TLG are small, thus less sampling sites (five sampling sites) were conducted. The detailed geographic information of sampling sites in the eleven mangrove regions was shown in Fig. S1 (Supplementary data, Fig. S1).

Surface sediments (0–5 cm) were collected for Hg analysis, and there were four replications for each sampling site. All sediment samples were sealed in polyethylene bags stored frozen in foam boxes. The samples were transported immediately to the laboratory and frozen at −20 °C. The samples were frozen-dried, grinded and pass 0.5 mm sieve before analysis. The soil was digested using 3:1 (V/V) mixture of HNO_3 and H_2SO_4 at 95 °C, and the concentrations of Hg in the digestions were determined using cold vapor atomic fluorescence spectroscopy (CVAFS). As for quality control, GBW07435 (reference materials), the blanks and blink duplicates were conducted to monitor the analytical quality. The mean value of the recovery rate in reference materials was 87.8%, and the relative differences were <10% for Hg in the duplicate samples.

Evaluation of Hg pollution status in the eleven mangroves along South China Coast

Two methods were employed to assess the status of Hg pollution. Single contamination index (P_i) is defined by the following equation: $P_i = \frac{C_i}{S_i}$ (Davaulter and Rognerud. 2001), where C_i means measured values of Hg, S_i means evaluation criterion of Hg. According to the first control standard of marine sediment quality in china (GB 18668-2002), $S_{Hg} = 0.2$ mg/kg. The P_i consequence can be described as follows: (1) $P_i \leq 1$, free pollution, level I; (2) $1 < P_i \leq 2$, slight pollution, level II; (3) $2 < P_i \leq 3$, mild pollution, level III; (4) $3 < P_i \leq 5$, moderate pollution, level IV; (5) $P_i > 5$, heavy pollution, level V.

Geo-accumulation index (I_{geo}) is an important index to evaluate contamination degree of single substances in sediment (Li et al. 2018; Men et al. 2018; Song et al. 2018). The formula of I_{geo} is defined as follow: $I_{\mathrm{geo}} - log_2 \frac{C_n}{1.5B_n}$ (Pathak et al. 2015), where C_n is actually examined Hg in the sediments, 1.5 is coefficient used to minimize the variation of background values. B_n is geochemical background concentration of Hg. The evaluation criterion of I_{geo} values

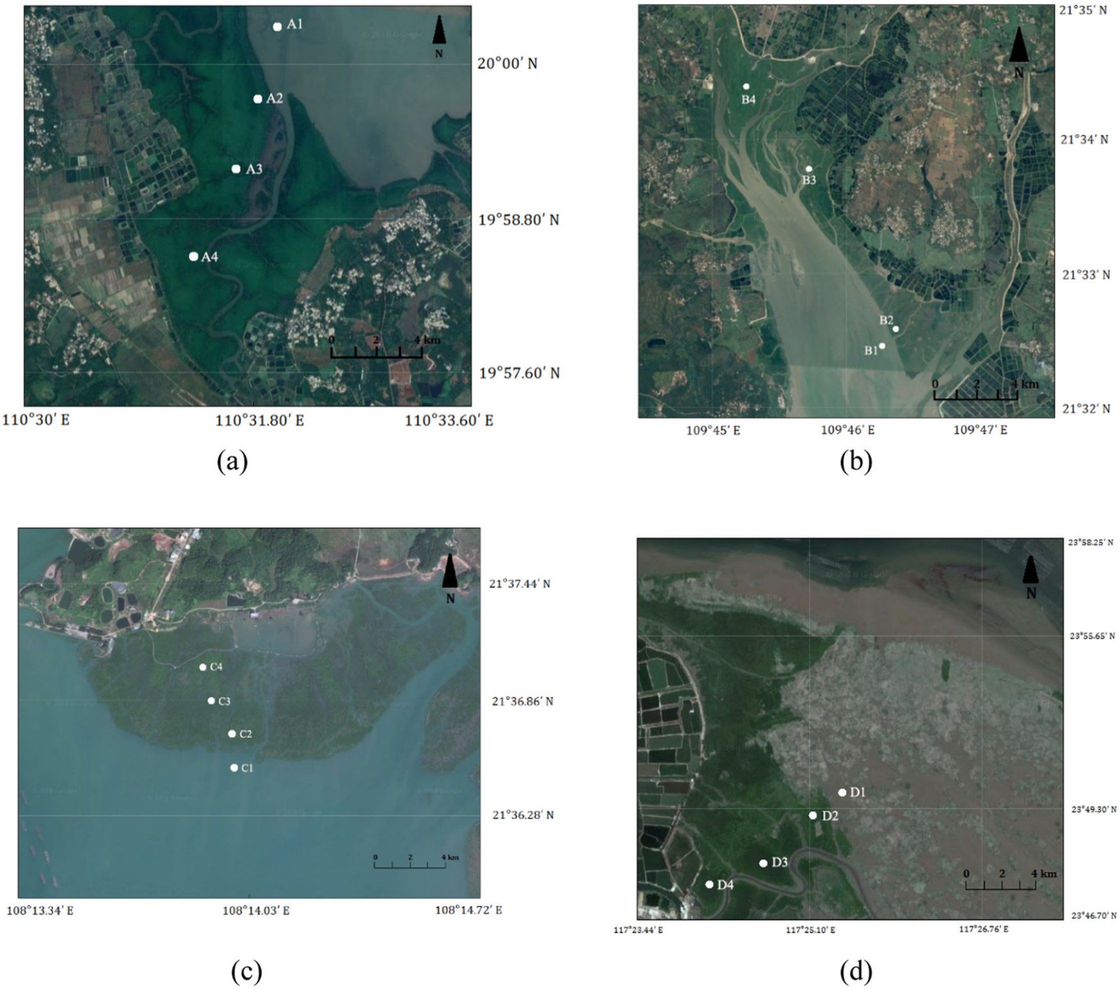

Fig. 1 Sampling sites in the four mangrove natural reserves with different tidal positions along tidal gradient (1 mudflat, 2 seaward mangrove, 3 medium mangrove and 4 landward mangrove), DZG (**a**), GQ (**b**), SJ (**c**) and ZJK (**d**)

is divided into seven classes: (1) $I_{geo} \leq 0$, practically uncontaminated; (2) $0 < I_{geo} \leq 1$, uncontaminated to moderately contaminated; (3) $1 < I_{geo} \leq 2$, moderately contaminated; (4) $2 < I_{geo} \leq 3$, moderately to heavily contaminated; (5) $3 < I_{geo} \leq 4$, heavily contaminated; (6) $4 < I_{geo} \leq 5$, heavily to extremely contaminated; (7) $I_{geo} > 5$, extremely contaminated.

Distribution of Hg in four selected mangrove regions along continuous intertidal gradient

Four mangrove natural reserves, namely DZG, GQ, SJ, and ZJK, were selected to investigate the distribution of Hg in the sediments along continuous intertidal gradient. The mangrove natural reserves in DZG, GQ, SJ, and ZJK are well protected, and exhibit obvious zonation along continuous tidal gradient from seaward to landward mangrove in the intertidal regions. Generally, pioneer mangroves are often found at the seaward front on the edge of mangrove forests, while typical Rhizophoraceous species always distributed in the middle and landward zones. On January 2017, the samples of sediments (0–5 cm) were collected. At each mangrove natural reserve, there were four tidal sampling sites (mudflat, seaward mangrove, medium mangrove, and landward mangrove), and in each tidal sampling site, four replications were conducted (Fig. 1). The measurements of Hg were measured as described above. Total organic carbon analyzer (TOC-L-CPH-SSM5000A) was used to determinate the contents of TOC in sediments.

Statistical analysis

Date of sediment Hg in each site were tested for their normality and variance with no transformation, and the data in each site were represented in mean ± standard deviation. The risks of Hg in the eleven mangrove regions were assessed basing on the individual statistical mean values, respectively. One-way ANOVA and least significant

Table 1 Comparisons of Hg concentrations (mg/kg) in sediments between China mangroves and other mangrove wetlands around the world reported in the literatures

Areas	Ranges of Hg concentrations	Hg mean concentrations	Sampling years	References
Mangroves, South China	0.0815–0.6377	0.2503	2017–2018	This research
Dongzhaigang, China	0.0077–0.9036	0.3141	2006–2007	Ding et al. (2009)
Fugong, China	0.113–0.135	0.1240	2013	Liang et al. (2013)
Futian, China	0.14–0.17	0.155	2011	He et al. (2014)
Sundarban Wetland, India	0.15–0.24	0.20	–	Chowdhury et al. (2017)
Sunderban Wetland, India	0.130–1.120	0.6525	2006	Sarkar et al. (2008)
South Gujarat Coast, India	0–15	–	2015	Dudani et al. (2017)
Port Klang, Malaysia	0.0004–0.2126	0.0247	2010	Haris and Aris (2013)
Strait of Malacca, Malaysia	–	0.0655	2013	Looi et al. (2016)
Eastern Tampa Bay, Florida, USA	0.0013–0.0830	0.027	2010–2011	Lewis and Russell (2015)
Sepetiba Bay, Brazil	0.006–0.092	0.03	–	Fonseca et al. (2013)
Guaratuba Bay, Brazil	0.0029–0.0442	0.0364	2004	Sanders et al. (2008)
North coast of Villa Clara, Cuba	0.032–1.81	0.3840	2009–2010	Olivares-Rieumont et al. (2012)
Fadiouth, Senegal, West Africa	0.005–0.013	0.010	2007–2008	Bodin et al. (2013)
Tanzanian coast, Tanzania, Africa	0.04–0.19	0.08	2014	Rumisha et al. (2016)
Coffs Harbor, Australia	–	0.023	–	Conrad et al. (2017)

difference (LSD) test was used to indicate the differences of Hg/TOC among different tidal positions along tidal gradient ($P < 0.05$).

Results

Concentrations of Hg in surface sediments collected from the eleven mangrove regions

Concentrations of Hg in surface sediments collected from eleven mangrove regions were shown in Table 1 and Fig. 2. Results showed that concentrations of Hg in the sediments collected from eleven mangrove regions ranged from 0.0815 to 0.6377 mg/kg, with an arithmetic mean value of 0.2503 mg/kg. For all the eleven regions, the measured Hg were more or less higher than their respective background values. The average concentrations of Hg in the eleven studied regions was: NS (0.4800 mg/kg), DZG (0.3395 mg/kg), QZ (0.3048 mg/kg), SY (0.2667 mg/kg), ND (0.2574 mg/kg), GQ (0.2466 mg/kg), TLG (0.2236 mg/kg), BMW (0.1843 mg/kg), SJ (0.1686 mg/kg), ZJK (0.1566 mg/kg), BLHK (0.1248 mg/kg), respectively.

Evaluation of Hg pollution in the eleven mangroves along South China Coast

The assessment values of P_i and I_{geo} were shown in Table 2. The contamination index (P_i) showed mild pollution ($2 < P_i < 3$) in NS; slight pollutions ($1 < P_i < 2$) in DZG, QZ, SY, ND, GQ, TLG, and free pollutions ($P_i < 1$) in BMW, SJ, ZJK and BLHK. The data of geo-accumulation index (I_{geo}) indicated NS, DZG, and SY were moderately to heavily contaminated ($2 < I_{geo} < 3$), TLG, BMW, QZ, and ND were moderately contaminated ($1 < I_{geo} < 2$), and GQ, ZJK, SJ, and BLHK were uncontaminated to moderately contaminated ($0 < I_{geo} < 1$) or practically uncontaminated ($I_{geo} < 0$).

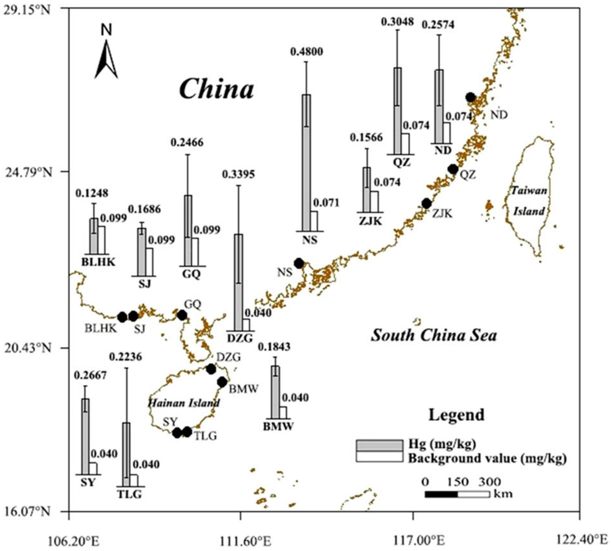

Fig. 2 Measured Hg and respective background Hg in the eleven mangrove regions

Distribution of Hg in the four selected mangrove reserves with complete mangrove zonation along tidal gradient

The trends of Hg and TOC in the sediments along tidal gradient from seaward mudflats to landward mangroves were shown in Fig. 3 and Fig. 4, respectively. For all the four natural reserves, concentrations of Hg in the sediments increased sharply form from seaward mudflats to landward mangroves. Increasing trends of soil TOC were also

Table 2 The values of contamination index in the eleven mangrove regions

Index	DZG	BMW	TLG	SY	GQ	BLHK	SJ	ZJK	QZ	ND	NS
P_i	1.698	0.922	1.118	1.334	1.233	0.624	0.843	0.783	1.524	1.287	2.400
I_{geo}	2.500	1.620	1.898	2.1522	0.7317	−0.2508	0.1831	0.4965	1.4573	1.2134	2.1722

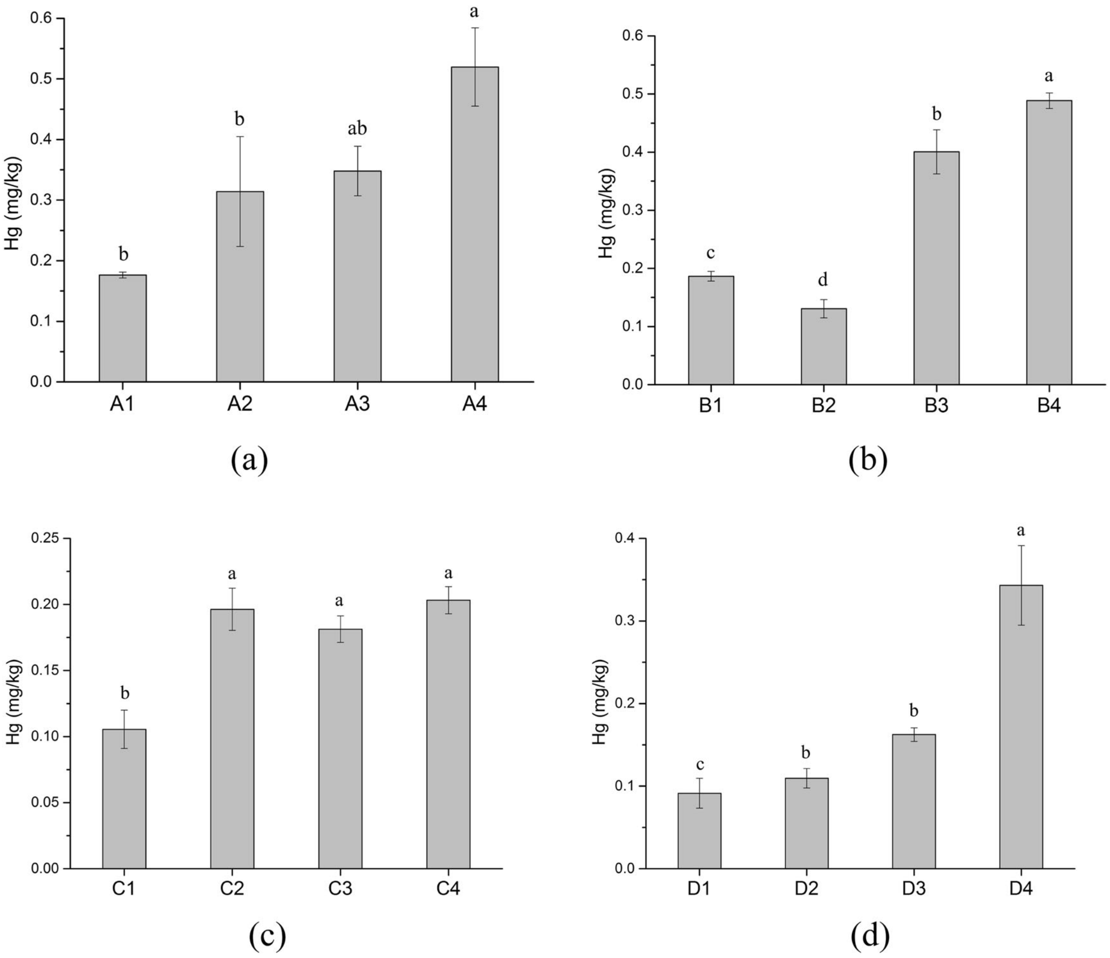

Fig. 3 Hg concentrations in the sediments along tidal gradient from seaward mudflats to landward mangroves in DZG (**a**), GQ (**b**), SJ (**c**) and ZJK (**d**). Different letters above the bars indicate significant differences among different tidal positions at $P < 0.05$ as determined by LSD test

observed with the increase of tidal gradient. Moreover, the data of Fig. 5 clearly illustrated a significant positive relation between Hg and TOC ($Y = 0.01693 \times -0.022$, $r = 0.688$, $P < 0.01$).

Discussion

The status of Hg contaminations in mangroves along South China Coast

The present data showed that the concentrations of Hg in the sediments of mangroves along South China Coast ranged from 0.0815 to 0.6377 mg/kg, approximately consisting with the previous reports (Ding et al. 2009; Vane et al. 2009; Liang et al. 2013; He et al. 2014). In terms of the comparisons of the present data and other mangroves around the world, the concentrations of Hg in mangroves along South China Coast were comparable or lower than those in India and Cuba (Haris and Aris 2013; Sarkar et al. 2008; Olivares-Rieumont et al. 2012; Looi et al. 2016; Dudani et al. 2017), but seemed to be higher than African mangroves (Table 1) (Bodin et al. 2013; Lewis and Russell 2015).

In this study, two methods were employed to assess the risk of Hg. The index of P_i mainly aimed to evaluate the toxicity risk of Hg based on absolute concentration of measured Hg, whereas NS scored the highest value, and the order of P_i in the eleven studied mangrove regions was: NS > DZG > QZ > SY > ND > GQ > TLG > BMW > SJ >

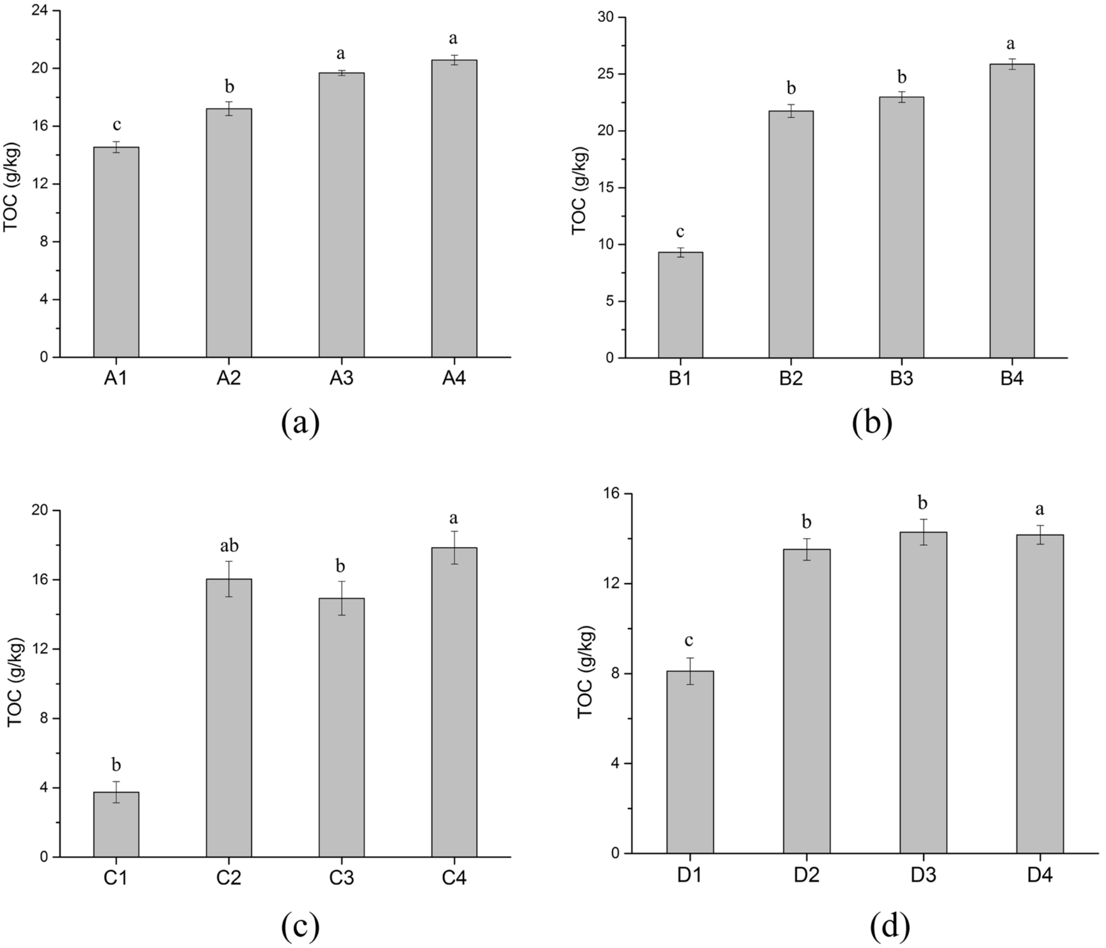

Fig. 4 Trends of TOC in the sediments along tidal gradient from seaward mudflats to landward mangroves in DZG (**a**), GQ (**b**), SJ (**c**) and ZJK (**d**). Different letters above the bars indicate significant differences among different tidal positions at $P < 0.05$ as determined by LSD test

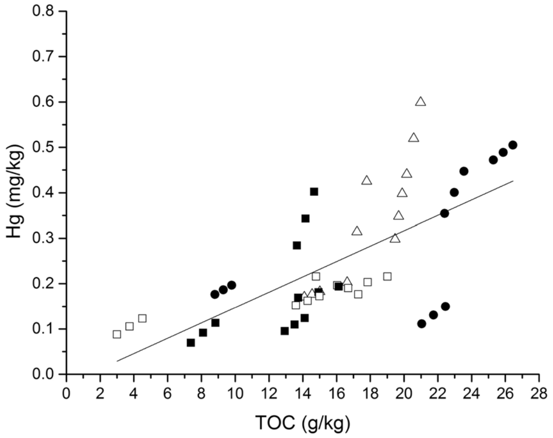

Fig. 5 The relationships between TOC and Hg concentrations along the continuous intertidal gradient from seaward mudflat to landward mangroves. DZG; GQ; SJ; ZJK

ZJK > BLHK. The highest sediment Hg in NS may partly ascribed to its special location, the center of Greater Bay Area, where possessed the highest population and most developed industry and economy in South China. The assessment of I_{geo} mainly focused on the relations between measured Hg and background Hg, the potential biological toxicity was ignored. The data of I_{geo} showed that NS, DZG, and SY exhibited that highest values, indicating moderate to heavy pollution inputs in these regions, TLG, BMW, QZ, and ND were considered as moderately contaminated. Both the results of P_i and I_{geo} indicated that the status of Hg in mangroves along South China Coast may strongly correlated to human activities, such as the distance from city, population density and economic level. The regions (SJ and BLHK) where far away from city and with lower GDP, consistently exhibited relative lower absolute Hg content and less pollution inputs.

Distribution of Hg in the sediments of mangroves along tidal gradient

In the four selected typical mangrove reserves, higher Hg was observed in the sediments of mangroves when compared to mudflat. The present data coincided with the issue that mangroves are potential reservoirs of pollutants (e.g., heavy metals and organic contaminants). Developed root systems would slow the flow of water during tidal cycle, leading to a rapid particle sedimentation and pollutant deposition (Li et al. 2015; Atwood et al. 2017). Lacking of interception mangroves, mudflat therefore may be more difficult in particle deposition and Hg accumulation. Besides, the presence of mangrove plants could change the textures of sediments (Wang et al. 2013; Tu et al. 2015). Hg is harder absorbed by the sediments with high ratio of sandy particles. Generally, the presence of mangrove could directly promote the ratio of clay, leading to a higher efficiency in nutrient enrichment and pollutant accumulation (Wang et al. 2013; He et al. 2014; Chen et al. 2015).

Moreover, the present data clearly indicated an increasing trendy of sediment Hg in mangroves with the increases of tidal gradient. Developed mangroves often show an interesting zonation in the intertidal regions. Similar phenomena was also observed in the four selected studied mangrove reserves, with a continuous succession from seaward pioneer species (*Avicennia marina* or *Aegiceras corniculatum*) evolved to the transitional and landward Rhizophoraceae species (*Kandelia obovata, Bruguiera gymnorrhiza and Rhizophora stylosa*). Generally, higher community diversity and productivity were often found in the stable community with latter succession. As for mangrove succession and zonation in South China, Rhizophoraceae species are much higher and bigger than pioneer mangrove species. In this study, we also found that plant species diversity was much higher in the landward Rhizophoraceae community (data not shown). The higher concentrations of TOC in landward mangroves may partly ascribed to their relative higher productivity and litter decomposition (Li et al. 2016). It has been reported that sediments with high TOC contents often possess higher adsorption capacity of heavy metals (Contreras et al. 2018; Wu et al. 2018). Significantly positive correlation between TOC and Hg was also observed. Besides, most pollutants are land-base sources, driving from municipal waste, fishponds and farmlands surrounding mangroves (Tam and Wong 2000; Feng et al. 2017).

Recently, organic mercury (e.g., methyl mercury, MeHg) has aroused more and more attention due to its high toxicity and potential risk (Gilmour et al. 1992; Bjørklund et al. 2017; Crowe et al. 2017). The formation of MeHg was regulated by soil property, methylated genes and microorganisms (Gilmour et al. 1992; Andrić et al. 2016; Bjørklund et al. 2017; Crowe et al. 2017). Unfortunately, dynamics of MeHg were not detected in the present study, more detailed information focused on dynamics of Hg and the risk assessments of MeHg in mangroves should be further conducted.

Conclusion

Results showed that concentrations of Hg in the sediments collected from eleven mangrove regions ranged from 0.0815 to 0.6377 mg/kg, with an arithmetic mean value of 0.2503 mg/kg. NS scored the highest value of P_i, and the order of P_i in the eleven studied mangrove regions was: NS > DZG > QZ > SY > ND > GQ > TLG > BMW > SJ > ZJK > BLHK. The data of I_{geo} showed that NS, DZG, and SY exhibited that highest values, indicating moderate to heavy pollution inputs in these regions. As for the distribution of Hg in the sediments along tidal gradient, a significant positive relation was observed between sediment Hg and TOC, the total Hg in sediments sharply increased from seaward mudflats to landward mangroves, coinciding with the increases of sediment TOC. The present data indicated that mangroves are potential reservoirs of pollutants. However, ecological risks of Hg, especially in some mangrove regions with high human activities, should be taken into special consideration in future mangrove management.

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Compliance with ethical standards

Conflict of interest The authors declare that they have no conflict of interest.

Ethical approval All the related authors confirmed there were no conflict of ethical approval.

Informed consent All the related authors have known the informed consent.

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