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Distribution of mercury in sediments, plant and animal tissues in Matang Mangrove Forest Reserve, Malaysia

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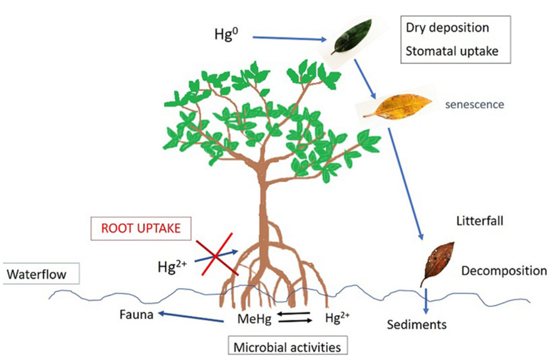
GRAPHICAL ABSTRACT



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ABSTRACT

Charcoal production activities at the Matang Mangrove Forest Reserve (MMFR) in Peninsular Malaysia have a potential to emit volatile compounds such as Hg back into the ambient environment, raising concerns on the public health and safety. The present study was aimed at analyzing Hg concentration from different plant/animal tissues and sediment samples (in total 786 samples) to understand clearly the Hg distribution at the MMFR. Leaves of *Rhizophora* spp. showed higher Hg concentration with an increasing trend from young, to mature, to senescent and decomposing stages, which was possibly due to accumulation of Hg over time. The low Hg concentration in *Rhizophora* roots and bark suggests a limited absorption from the sediments and a meagre accumulation/partitioning by the plant tissue, respectively. In the case of mangrove cockles the concentration of Hg was lower than the permissible limits for seafood consumption. Although the mangrove gastropod - *Cassidula aurisfelis* Bruguière had rather elevated Hg in the muscle tissue, it is still less than the environmental safely limit.

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Beside the chances of atmospheric deposition for Hg, the sediment samples were found to be unpolluted in nature, indicating that in general the MMFR is still safe in terms of Hg pollution.

1. Introduction

Mercury (Hg) is an element of great concern because of its high toxicity and long persistence in the environment (Pirrone et al., 2010). Presence of Hg in the environment can be due to natural or anthropogenic causes. The natural sources of Hg include volcanic eruptions, weathering processes of the Earth's crust and forest fires (Fitzgerald et al., 1998), whereas anthropogenic sources are coal-fired power plants (Habuer et al., 2016), agricultural chemicals (*e.g.* fungicides and pesticides), mineral mining, industrial discharge, medical waste incineration and municipal waste combustion (Pirrone et al., 2010; Sloss, 2012). Meanwhile, industrial expansion and settlements in the coastal areas are additional burdens of pollution nowadays (Lewis et al., 2011). Recently, atmospheric deposition (with a long-range transport) was recognized as a main route for Hg to enter aquatic ecosystems and thus become available in high concentrations in some remote areas and pristine habitats (Fitzgerald et al., 1998; Chen et al., 2014; Wiedinmyer et al., 2014). The chemical speciation and the mobility of Hg in the environment are also influenced by biotic and abiotic components and their characteristics. Therefore, it warrants a careful attention for understanding the direct input of Hg or relevant pathways in Hg cycling.

In the tropical and subtropical regions, mangroves not only act as important nurseries and feeding ground for various finfish and shellfish species but also as depositional areas for domestic/industrial pollution, including inorganic Hg from atmospheric deposition and runoff (Silva et al., 1990; MacFarlane et al., 2003; Analuddin et al., 2017). Microbial action in the mangrove areas, with anoxic sediments enriched with dissolved organic matter and humic acids (Ding et al., 2009), would be able to convert the inorganic Hg into methylmercury (Me-Hg), one of the most toxic forms (Hall et al., 2008). Under these circumstances, the Hg speciation through biomagnification from primary consumers to predators leads to several impacts on the dependent populations (Senn et al., 2010).

A great deal of studies on Hg in mangroves focused on pollution status, mostly by looking at the sediments (Lewis et al., 2011) and bioaccumulation in the food chain (Zuykov et al., 2013; Le et al., 2018). However, the scientific literature available on the partitioning of Hg within plant tissues is limited and still has to evaluate its intake/absorption mechanisms from the environment (Li et al., 2016). Regarding the accumulation of Hg in mangrove tissues, roots have a capacity to absorb aqueous Hg sources (Walsh et al., 1979), but translocation to other plant parts is not the same for all species due to different physiological mechanisms. For instance, salt excreting mechanisms present in some species could be associated to higher translocation factors than species that are salt-excluding or salt-accumulating (Ding et al., 2011). The concentration of Hg in leaves also depends on the air pollution level, as Hg can be absorbed directly from the atmosphere (Schroeder and Munthe, 1998). Inside the leaves, Hg tends to bind to -SH groups in amino acids, metallothioneins, glutathione or phytochelatins, the latter being specific metal-binging peptides with structure (ϒGluCys)n Gly (n = 2–11), that can detoxify trace metals (Huang and Wang, 2010). Juvenile mangroves can show the symptoms of Hg poisoning through loss of turgor, epinasty, leaf abscission, chlorosis and blackening of leaf and stem, if its concentration exceeds 500 mg Kg^{-1} in the soil (Walsh et al., 1979).When Hg replaces other metal ions in the photosynthetic machinery, there will be a decline in photosynthetic rate (Huang and Wang, 2010). Among plant tissues, bark can represent the sink of Hg through atmospheric deposition, surface absorption and binding to thiol groups and tannins inside the tissue (Serbula et al., 2012; Chiarantini et al., 2017; Chiarantini et al., 2016)

As plants are at the base of the trophic chain, it is fundamental to consider Hg concentration (hereafter referred to as [Hg]) in plants, together with sediment and associated fauna. Among the mangrove macro-benthic invertebrates, molluscs are well recognized for their economic potential and relationship with vegetation/edaphic gradients (Bosire et al., 2008; Kon et al., 2010; Chen and Ye, 2011; Andreetta

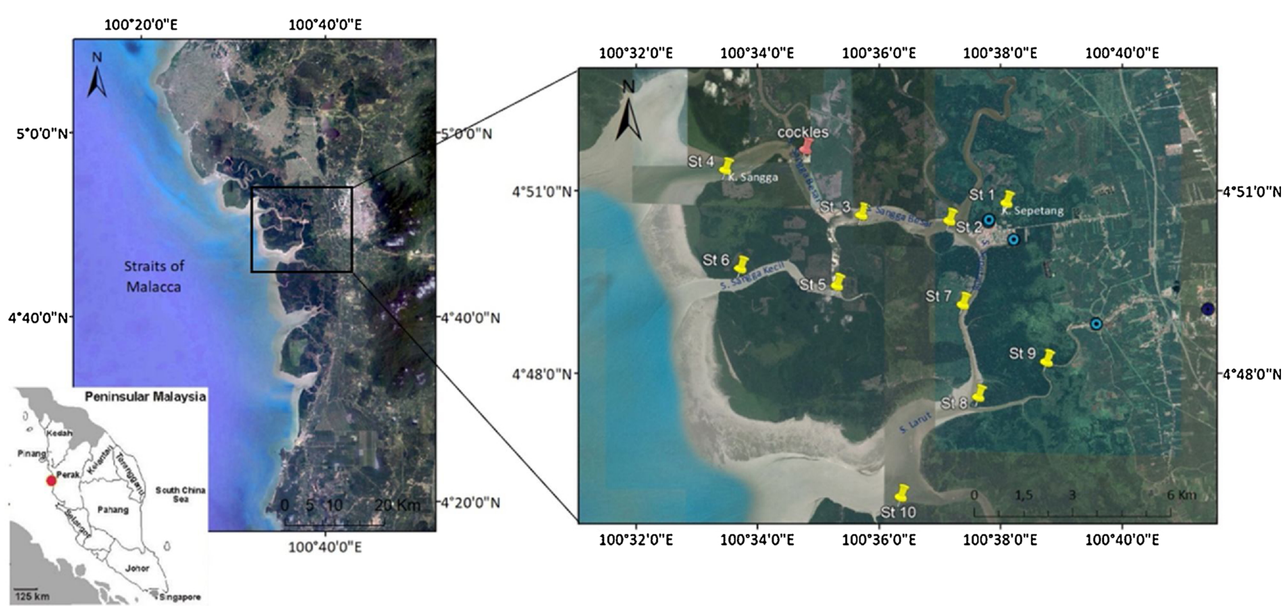

Fig. 1. Matang Mangrove Forest Reserve on the west coast of Peninsular Malaysia showing ten sampling stations (Yellow pinpoints St 1–10) for plant/animal tissues and sediment collection. Mangrove cockles were separately collected from a culture farm (red pinpoint). Location of charcoal kilns (light blue circles) and rubber glove factory (dark blue circle) are also marked. The sampling stations represent four spatial (land-sea) gradients - Gradient A with St 1–4, Gradient B with St 1–3 and 5–6, Gradient C with St 1,7–8 and 10, and Gradient D with St 8–10, for understanding the impact of local anthropogenic activities in relation to Hg pollution. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article).

et al., 2014). Nevertheless, these organisms are sensitive to the local environmental changes or pollution and hence are considered as bioindicators (Chen et al., 2007; Ellison, 2008; Cannicci et al., 2009; Penha-Lopes et al., 2010; Zvonareva et al., 2015).

The present study was conducted in Matang Mangrove Forest Reserve (MMFR) in Peninsular Malaysia where *Rhizophora apiculata* Blume and *R. mucronata* Lamk. are subjected to a 30-year forest rotation cycle for pole and charcoal production (Ariffin and Mustafa, 2013; Chong, 2006; Goessens et al., 2014). For the production of charcoal, there are as many as 489 igloo shaped kilns available in the vicinity. When the rate of charcoal production from each kiln *i.e.*, 10 − 11 t out of 40 t greenwood in a month is considered (Quispe -Zúñiga et al., 2014) then the yearly production of charcoal at Matang reaches up to 60,000 t. This year-round production of charcoal not only releases carbon, but also has a potential to emit volatile compounds like Hg back into the ambient environment. In fact, burning of charcoal is responsible for Hg emissions ten times higher than the burning of fossil fuels per unit of combustion, hence possibly representing a hazardous Hg source (Pandey et al., 2009). Other possible emission sources of Hg around Matang could be through industrial (rubber glove factory) discharge, use of chemicals in agriculture (palm oil) plantations, cargo ships and fishing vessels in the Malacca Strait, and a coal-fired power plant (TNB Janamanjung located 80 km south of MMFR) (Mokhtar et al., 2014).

With the raising concerns over human/ecosystem health on one hand and very limited (old) information about heavy metal pollution on the other, the present study observed [Hg] in both plant and animal tissues including surface sediment in the vicinity of MMFR. The objectives are (i) to find the partitioning of Hg in different tissues of *Rhizophora* spp. in relation to mangrove sediment and associated fauna (molluscs), (ii) to observe a Hg gradient, if any, away from its possible source(s) of emission on the landward side and, (iii) to assess the MMFR in terms of Hg pollution and environmental safety.

2. Materials and methods

2.1. Sampling area

The MMFR, located on the west coast of Peninsular Malaysia in the State of Perak, is extending over 40,466 ha (Jusoff and Taha, 2008). The entire reserve belongs to three administrative ranges - Kuala Sepetang, Kuala Trong and Sungai Kerang, with nearly 108 compartments of different aged *R. apiculata* and *R. mucronata* stands to support the ongoing (pole and charcoal production) management (Ariffin and Mustafa, 2013). The present study was conducted from Kuala Sepetang area where nearly 179 kilns with a yearly charcoal production capacity of 21,500 – 23,500 t are available (Fig. 1). Whether or not the century old MMFR management, which started in 1902 (Noakes, 1952), has been responsible for possible air and water quality deterioration in this area remained unanswered until the present study was initialized. To understand the impact of anthropogenic activities in the vicinity the sampling stations chosen addressed landward, midstream and seaward locations along the major water channels in the estuary in such a way that they represent four spatial (land-sea) gradients, namely Gradient A with St 1–4, Gradient B with St 1–3 and 5–6, Gradient C with St 1, 7–8 and 10, and Gradient D with St 8–10 (Fig. 1).

The mangroves at MMRF are regularly inundated by semidiurnal tides, with a mean spring high water of 2.65 m (NHC (National Hydrographic Centre of Malaysia), 2018). Mean annual rainfall is about 2109 mm, with two peaks in a year – one during April with the onset of southwest monsoon and another during November with northeast monsoon, respectively (Tanaka and Choo, 2000).

2.2. Sampling strategy

Surface sediments (uppermost 2 − 5 cm) from both riverbank and inside the forest (10 − 15 m from fringe) were collected from each station in five replicates (using a hand shovel) to assess the differences in Hg between open and understory (sediment) conditions. Since the mangrove surface sediments play a significant role for microbial activity, leaf decomposition, forage for gastropods, etc., our study did not focus on the difference in [Hg] with soil depth.

Plant tissue samples were collected from *R. apiculata* (in all sampling stations except St 4 and St 6) or *R. mucronata* (in St 4 and St 6 as *R. apiculata* was not present in these sites) (Table 1).

For mangrove leaves, ten replicates comprising 4–5 leaves each were collected randomly for young, mature, senescent and decomposing stages from ten different trees. Also, bark and root samples were collected randomly from six trees per each station. For fauna, the specimens of the widely distributed mangrove gastropod *Cassidula aurisfelis* Bruguière (Sasekumar and Chong, 1998) were collected in six replicates (Table 1) from the stations in which it was present (St 1–6). In addition, samples from the edible mangrove cockle *Tegillarca granosa* L. were collected in ten replicates (from a cockle culture farm located between St 3 and St 4) to assess the health risk for consumers. In total 786 samples were packed in plastic zip-lock bags and kept in an icebox until reaching the laboratory (at Universiti Malaysia Terengganu) for further preservation and analyses.

Table 1
Sample size in relation to sediment, plant animal tissues collected from the Matang Mangrove Forest Reserve. Units corresponds to one leaf for leaves, to one individual for molluscs. The replicates are the result of pooling of sample units to have enough material for the Hg analysis, and the values reported are per each station.

Sample	Position / part	Stations	Replicates	N° units per replicate
Sediment	River bank	1,2,3,4,5,6,7,8,9,10	5	–
	Inside forest	1,2,3,4,5,6,7,8,9,10	5	–
Rhizophora apiculata	Young leaves	1,2,3,5,7,8,9,10	10	5
	Mature leaves	1,2,3,5,7,8,9,10	10	5
	Senescent leaves	1,2,3,5,7,8,9,10	10	5
	Decomposing leaves	1,2,3,5,7,8,9,10	10	5
	Bark	1,2,3,5,7,8,9,10	6	1
	Roots	1,2,3,5,7,8,9,10	6	1
R. mucronata	Young leaves	4.6	10	5
	Mature leaves	4.6	10	5
	Senescent leaves	4.6	10	5
	Decomposing leaves	4.6	10	5
	Bark	4.6	6	1
	Roots	4.6	6	1
Cassidula aurisfelis	Individuals	1,2,3,4,5	6	3
	Individuals	6	7	3
Tegillarca granosa	Individuals	Cockle culture farm	10	2

2.3. Sample preparation and Hg measurements

All samples, except sediments, were carefully washed with distilled milliQ water (Millipore Corporation, USA). Both biotic and sediment samples were subsequently frozen at −80 °C for 48 h and dried in a freeze dryer (LABCONCO Freeze Dry System/ Freezezone 4.5) at −40 °C with pressure lower than 0.133 mBar for 48 − 72 h. Before drying, the root specimens were separated into epidermis, cortex and xylem tissues. Also, the muscle tissues of mollusks were gently dissected and their wet weight was measured (SARTORIUS CP224S, 0.1 mg precision). After oven drying, the muscle tissues were once again weighed for their dry-weight to estimate the moisture content used to calculate the [Hg] on wet- weight. All samples were then ground to a fine powder using mortar and pestle. Sediment samples were sieved with a 60 μm mesh size.

The [Hg] in powdered samples was measured using a cold vapor atomic absorption spectrometer (MA-3000; Nippon Instruments Corp., Japan) with detection limit of 0.002 ng. All chemicals used were of analytical grade and the reagents were prepared as per the instructions in NIC-600-2166-03 (Nippon Instruments Corp.). A calibration curve to estimate the [Hg] (mg Kg^{-1} dry weight) in each sample was generated from 0, 0.1, 0.5 and 1.0 ppm standards.

2.4. Accuracy assessment

The accuracy of Hg measurements in samples was assured through certified reference materials (CRMs). For plant tissues and mollusks, the CRM NIST-SRM2976 (freeze-dried mussel tissue) with a concentration of 61.0 (±3.6) μg Kg^{-1} was chosen, whereas for sediments the CRM NIST-SRM2702 (marine sediments) with a concentration of 447.4 (±6.9) μg Kg^{-1} was used. The measurements for the CRMs were taken at the first and last sample in every sequence that totaling to 6 times for the SRM2702 and 15 times for the SRM2976. The percentages of recovery were 81.9–104.8% for SRM2976 and 75.4–127.1% for SRM2702.

2.5. Geo-accumulation index

Geo-accumulation index of sediments (Muller, 1979) was calculated from the equation given below:

$$\mathrm{Igeo} = \log 2 \frac{[\mathrm{Hg}]_{\mathrm{S}}}{1.5 \times [\mathrm{Hg}]_{\mathrm{bl}}} \quad (1)$$

where, I_{geo} is geo-accumulation index, [Hg] is mercury concentration in sediments (s) and background level for sediments (bl). Factor 1.5 is the background matrix correction factor, including the variation of background levels due to lithogenic effects (Muller, 1979).

The background level of Hg for west coast of Peninsular Malaysia was considered equal to 30 μg Kg^{-1} (*cf.* Looi et al., 2015). Based on geo-accumulation index values, the sediment pollution status can be divided into 6 categories - unpolluted (0–1), moderately to unpolluted (1–2), moderately polluted (2–3), moderately to highly polluted (3–4), highly polluted (4–5), and very highly polluted (> 5) (Chowdhury et al., 2017).

2.6. Statistical analyses

Statistical analysis was performed using R software to find the significant differences in Hg between samples and stations. Prior to analysis, all data were checked for the normality using Shapiro-Wilk and Levene's tests. Non-parametric tests like Kruskal-Wallis was used for the data displaying non-normality.

3. Results

3.1. Mercury in sediments

The sediment samples collected from the riverbank and inside the forest did not differ significantly for [Hg], except in St 2 and St 10 (Fig. 2). In relation to the four spatial gradients, St 2 was common to gradients A and B and St 10 to gradients C and D. At St 2 the [Hg] was found to be higher in the riverbank sediment (average, 79.2 ± 18.6 μg Kg^{-1}) than in the forest sediments, whereas at St 10 it was higher towards the forest interior (73.9 ± 11.2 μg Kg^{-1}). If the seaward locations are considered (*i.e.* St 4, 6 and 10), [Hg] at St 10 for both riverbank and inside forest sediments was significantly higher than in St 4 (riverbank: 34.0 ± 2.6 and inside forest: 39.9 ± 4.1 μg Kg^{-1}) and St 6 (riverbank: 36.1 ± 1.0 and inside forest: 38.1 ± 3.5 μg Kg^{-1}) (Kruskal-Wallis test, $p < 0.05$) (Fig. 2). However, the geo-accumulation index of Hg was found to be < 1 for all sediment samples (Table 2).

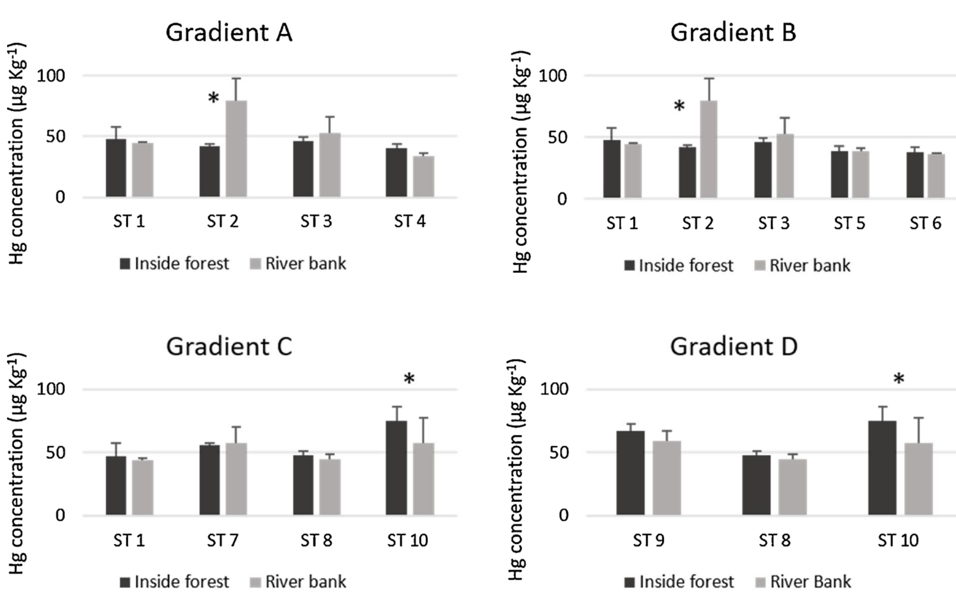

Fig. 2. Concentration of Hg in mangrove sediment in relation to the four land-sea gradients established for Matang Mangrove Forest Reserve (see Fig. 1 for A–D gradient stations). Statistical differences between riverbank and inside forest sediments were marked by * (Kruskal-Wallis test, $p < 0.05$).

Table 2
Geo-accumulation index of Hg in the mangrove sediments collected from Matang Mangrove Forest Reserve.

Geoaccumulation index		
Station	Sediments inside forest	Sediments riverbank
St 1	0.07	−0.03
St 2	−0.09	0.82
St 3	−0.02	0.23
St 4	−0.17	−0.41
St 5	−0.22	−0.22
St 6	−0.24	−0.32
St 7	0.32	0.36
St 8	0.10	−0.01
St 9	0.49	0.25
St 10	0.72	0.27

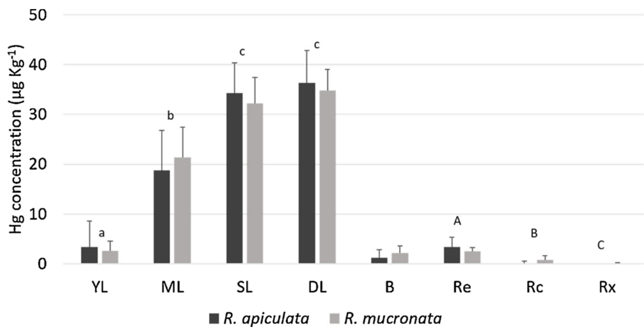

Fig. 3. Comparison of Hg concentration among plant tissues in Matang Mangrove Forest Reserve - young leaves (YL), mature leaves (ML), senescent leaves (SL), decomposing leaves (DL), bark (B), root epidermis (Re), cortex (Rc) and xylem (Rx) of *Rhizophora apiculata* and *R. mucronata*. Average values from all stations were pooled and represented with standard deviation in the graph. While capital alphabets above each bar indicate significant differences (Kruskal-Wallis test, $p < 0.05$) among root tissues, the small alphabets are for different leaf stages.

3.2. Mercury in plant tissues

No statistical differences in [Hg] were observed between *R. apiculata* and *R. mucronata* at the MMFR (Fig. 3). For leaves, there was a significant increasing trend of [Hg] from young to mature to senescent and decomposing stages in the order. While young leaves showed 3.3 ± 5.3 $\mu g\ Kg^{-1}$ of [Hg], the senescent leaves had 34.3 ± 6.0 $\mu g\ Kg^{-1}$ in *R. apiculata* (Kruskal-Wallis test, p < 0.05). The variation of [Hg] between senescent and decomposing leaves was non-significant (Kruskal-Wallis test, p > 0.05).

The [Hg] in root epidermis was 3.3 ± 2.0 $\mu g\ Kg^{-1}$ for *R. apiculata* and 2.5 ± 0.8 $\mu g\ Kg^{-1}$ for *R. mucronata*, yet lower than the leaves. However, there was a significant decrease in [Hg] from root epidermis on the outside to cortex and xylem inside (Kruskal-Wallis test, p < 0.05) (Fig. 3). For the bark, [Hg] was found to be 1.2 ± 1.6 $\mu g\ Kg^{-1}$ for *R. apiculata* and 2.0 ± 1.4 $\mu g\ Kg^{-1}$ for *R. mucronata* (Fig. 3). For mature leaves, there was a general tendency of increasing [Hg] from land to sea, except for St 6 in Gradient B (Fig. 4) (Kruskal-Wallis test, p < 0.05). No such spatial variations were observed for any other plant tissues analyzed.

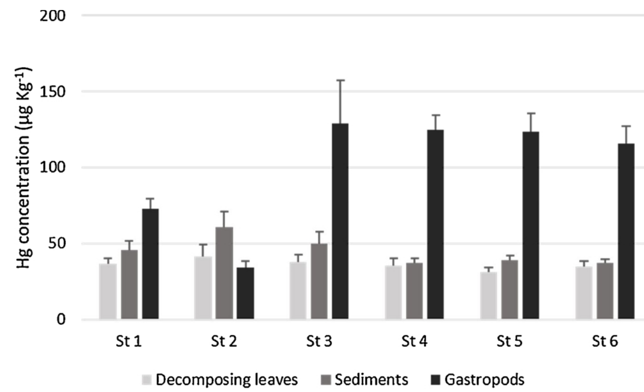

Fig. 5. Comparison of Hg concentrations among sediments (average of riverbank and forest sediments), decomposing *Rhizophora* leaves and gastropods (*Cassidula aurisfelis*) collected from Matang Mangrove Forest Reserve. Data presented were mean values and standard deviation.

3.3. Mercury in mollusc tissues

On one hand, the mangrove gastropod *C. aurisfelis* showed higher [Hg] than the sediment or plant tissues for most stations (Fig. 5), especially in the locations away from landward margins with a significant spatial difference (Table 3). On the other hand, the mangrove cockle *T. granosa* had much lower Hg concentration (27.6 ± 4.2 $\mu g\ Kg^{-}$

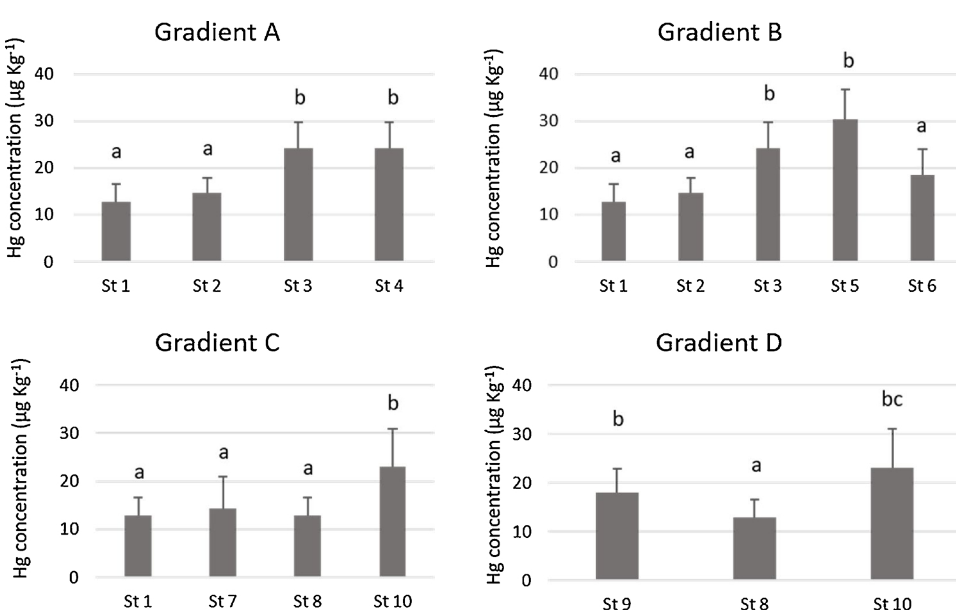

Fig. 4. Distribution of Hg in mature leaves of *Rizophora* spp. along four land-sea gradients established for Matang Mangrove Forest Reserve (see Fig. 1 for A–D gradient stations). Data presented were mean values and standard deviation. Statistical differences among the stations are marked by small alphabets (Kruskal-Wallis test, $p < 0.05$).

Table 3
Hg concentrations in mangrove gastropod - *Cassidula aurisfelis*, collected from Matang Mangrove Forest Reserve. The values correspond to both dry weight (dwt) and wet weight (wwt). Statistically differences (Kruskal-Wallis test, p = 0.0001) among the dry weight measurements are marked by small alphabets.

Stations	Hg dwt (μg Kg^{-1})	Hg wwt (μg Kg^{-1})
St 1	73.0 ± 6.5 [a]	7.8 ± 1.8
St 2	34.1 ± 4.5 [a]	3.9 ± 0.6
St 3	128.7 ± 28.6[b]	14.9 ± 2.8
St 4	125.0 ± 9.3[b]	17.0 ± 2.4
St 5	123.6 ± 12.2[b]	18.9 ± 1.8
St 6	115.7 ± 11.4[b]	17.3 ± 2.0

[1] dry weight and 21.7 ± 3.1 μg Kg^{-1} wet weight) compared to the gastropods at Matang.

4. Discussion

4.1. Sediments

Higher [Hg] values at both St 2 and St 10 could be linked to the presence of charcoal fragments in the sediments (visual observation). At St 10 - being located close to the 'productive' forest (compartment #49) (Ariffin and Mustafa, 2013), the tree cutters live temporarily and use charcoal for chargrilling fish and crabs (for daily cooking they use liquefied petroleum gas). Leftover fragments of charcoal might have percolated into the sediment and led to the observed higher [Hg] in this station. In fact, charcoal can easily absorb Hg compared to normal sediments (Madren, 2014), behaving similar to activated carbon (Tan et al., 2011). However, in the case of St 2, the forest belongs to the 'protective' category and known as the Virgin Jungle Reserve (VJR). As there is no scope for human intervention here, the upstream water flow and transportation from the nearby charcoal factories may have brought the charcoal fragments. For St 10, receiving pollutants from a rubber glove factory and oil palm plantations upstream is also plausible. The [Hg] at the mouth of River Larut (St 10) was significantly higher than that at the mouth of River Sangga Besar (St 4) or River Sangga Kecil (St 6).

The Hg levels in sediments could also vary depending on the exposure of mudflats to tidal washing and vegetation cover, which emphasizes the role of mangrove roots in trapping sediments, leaf litter as well as Hg (Correia and Guimarães, 2016; Le et al., 2017). In the present study, the non-significant differences of [Hg] between riverbank and inside forest sediments for the majority of sites could be linked to the presence of homogenous vegetation, with small differences of canopy cover, and root presence from fringe line to the forest (10 − 15 m) inside.

The [Hg] in MMFR sediments was however lower than the threshold value of 130 μg Kg^{-1} and the probable effect level of 700 μg Kg^{-1} reported by MacDonald et al. (1996) (Macdonald et al., 1996). In fact, the other mangrove areas like Port Klang in Peninsular Malaysia (Haris et al., 2017), Futian in China (Niu et al., 2019), and South-East Sulawesi in Indonesia (Analuddin et al., 2017) recorded higher [Hg] than at Matang (Table 4). In light of the background level of Hg reported for Malacca Straight (Looi et al., 2015) and the present geo-accumulation index values, the sediment samples collected from MMFR are still indicative of a non-polluted nature.

4.2. Plants

Plants are characterized by different concentrations of trace metals in their body, mostly in relation to their physiology. For instance, the absorption of metals by root tissues can be linked to conditions such as the bioavailability of elements in the soil (Kumar et al., 2017), the capacity of roots to absorb the elements, etc. Some species have a capacity to translocate the metals from the roots to other parts like bark and leaves, while others follow exclusion mechanisms (Weis and Weis, 2004).

In the case of *Rhizophora* spp. at Matang, the low [Hg] in roots was possibly due to unpolluted state of the sediments and very low uptake by roots because of Hg binding to sulfides and organic matter (Niu et al., 2019). However, Walsh et al. (1979) found a higher absorption of Hg by *R. mangle* seedlings. In this context, further evaluation on the metal absorption rates among juvenile, young and adult stages of different mangrove species is required, not only for Hg, but ideally for a wide suite of heavy metals. Higher [Hg] in *Rhizophora* leaves compared to roots clearly suggests that translocation from its root system to be less and uptake from the air or atmospheric deposition to be more. This was further supported by the increasing order of [Hg] from young to mature to senescent stages and its accumulation over time in the leaf tissues. Similar results were also found by Ding et al. (2011) (Ding et al., 2009) in mangroves as well as terrestrial plants in China, where they indicated that Hg^{2+} is rather immobile in the leaf tissue (Gaggi et al., 1991; Ericksen et al., 2003; Laacouri et al., 2013). However, low Hg levels in the mangrove bark at Matang, in contrast to other locations elsewhere, *e.g.* Sundarbans in India (Muller, 1979), could be due to prevailing interspecies, regional, individual and tissue differences (Ding et al., 2009). The canopy structure at managed mangrove sites in MMFR is quite uniform as all trees have the same age and the forest gets a closed canopy after 6 years on average (Otero et al., 2019), while in the Sundarban mangroves the forest structure is much more variable and therefore stems are more exposed to the atmosphere and to Hg deposition. Moreover, Chowdhury et al. (2017) have linked the accumulation of Hg in *R. apiculata* bark with the atmospheric deposition due to burning of fossil fuel and garbage by local people along with agricultural and chemical run-off. No doubt the bark is useful for biomonitoring of airborne Hg pollution (Chiarantini et al., 2016), but it is uncertain for mangroves - at least for the observations made in the present study- and, leaves may be a more suitable indicator of Hg in plant tissues. Perhaps future studies focusing on the degree of tree bark exposure to different atmospheric variables (with open/closed canopy from waterfront to back mangroves in different seasons) would be able to provide more scientific insights.

4.3. Molluscs

Bioaccumulation and trophic transfer processes from the primary producers to the primary consumers (Senn et al., 2010) seemed to be important for the higher [Hg] found in *C. aurisfelis* at Matang. Earlier,

Table 4
Range of Hg concentration in mangrove sediments from the present study and other studies from different areas.

Country	Hg concentration (μg Kg-1)	Reference
Malaysia - MMFR	36.1 - 79.2	Present study
Malaysia - Port Klang	0.01-150	Haris et al., 2107 ()
China	151-172	Li et al., 2016 (Li et al., 2016)
China	26.1-438	Ding et al., 2011 (Ding et al., 2009)
China	154.7 - 218.4	Niu et al., 2018 (Haris et al., 2017)
Indonesia	190-280	Analuddin et al., 2017 (Analuddin et al., 2017)

Le et al. (2017) also observed similar range of [Hg] in the mangrove gastropods collected from Setiu wetlands in Peninsular Malaysia. However, the reason for lower [Hg] in *C. aurisfelis* at landward sites (St 1 and St 2) in contrast to the sediments and decomposing leaves on the forest floor (Fig. 5) is rather unclear but may be dependent on the Hg forms available, especially the organic mercury that is readily absorbed by the animals (Costa et al., 2012).

Lower [Hg] in mangrove cockles compared to gastropods fetches the idea of bioaccumulation difference in pelagic and benthic feeders, respectively. *Cassidula aurisfelis* is a herbivore (Costa et al., 2012) that feeds mostly on mangrove leaf litter and grazes organic matter on the surface sediment, whereas *T. granosa* is a filter feeder that is exposed to Hg from diet derived from phytoplankton and particulate organic matter in the water column (Costa et al., 2012). Bioavailability of Hg in the water is usually less than in the sediments under forest cover, which has a higher organic matter content (Le et al., 2017; Pant and Allen, 2007). Furthermore, the higher [Hg] in gastropods and mature leaves at the seaside, compared to the landward sites, might suggest an influence of intense cargo shipping and fishing vessel activities in the Malacca Strait, known to be one of the world's busiest maritime routes (Ismail et al., 2016). However, further evidence is needed on this matter in order to be conclusive.

According to the commission regulation no. 466/2001 (EC, 2000), the permissible limit of Hg for seafood consumption is 500 µg Kg^{-1} (on wet weight). So, the lower [Hg] in *T. granosa* implies no health hazards for its consumption at Matang. In fact, Kuala Sepetang in the MMFR is well-known for the best cockle production and export to other locations in Malaysia (Mr. Lee, village head Kuala Sangga, pers. comm.). These commercially important mangrove cockles often draw a scientific attention due to increased pollution in the coastal habitats, especially in Asia (Rahayu et al., 2016; Sudsandee et al., 2017).

4.4. Mercury pathway in Matang

The limited accumulation of Hg in *Rhizophora* roots at Matang excludes aqueous root uptake and subsequent translocation to other plant parts as a main pathway. Higher Hg levels in the leaves suggest that litterfall can play an important role in Hg biogeochemical cycles here. In this context, Hg could partially be released back into atmosphere or converted into organic forms (*e.g.* methylmercury-MeHg) by microbes in the anoxic mangrove sediment enriched with dissolved organic matter (Correia and Guimarães, 2016).The Hg forms ultimately enter the food web through a trophic pathway, and in this case the molluscs being considered as primary consumers showed elevated Hg levels. Mercury can also be exported to adjacent habitats by physical factors such as tidal regimes and river flow (Bergamaschi et al., 2012) or trophic relay by migratory species (Hammerschmidt and Fitzgerald, 2006). Overall, the Hg pathway in Matang suggests the input of Hg be mainly from atmospheric mixed sources, without any specific connectivity to the charcoal and industrial activities in the vicinity.

5. Conclusions

The present study has examined the Hg concentrations in *Rhizophora* spp. at MMFR for the first time with a most reliable sample size. Among the biotic tissue samples, only leaves showed high [Hg] with an increasing trend from young to mature to senescent and decomposing stages that possibly associated with its accumulation over time. The pathway of mercury in *Rhizophora* suggests less chances of translocation from its root system and more chances for an uptake from the air or by atmospheric deposition. However, further evidences on the metal absorption rates among juvenile, young and adult stages of different mangrove species, degree of tree exposure to different atmospheric variables, etc., would be able to provide a better understanding on the mercury transfer at Matang. At present, the geo-accumulation index values are indicating unpolluted nature of the mangrove sediment at Matang. Although charcoal fragments in the sediment revealed higher [Hg], no clear spatial (land-sea) gradients could be inferred in relation to the charcoal factories. The low [Hg] in *T. granosa* imply no health hazards for its consumption. Overall, the MMFR is still safe in terms of Hg pollution but requires a timely check and assessment in light of the sustained charcoal production and other industrial developments in the vicinity.

CRediT authorship contribution statement

Giovanna Wolswijk: Data curation, Formal analysis, Funding acquisition, Investigation, Writing - original draft, Writing - review & editing. **Behara Satyanarayana:** Conceptualization, Funding acquisition, Resources, Supervision, Validation, Writing - review & editing. **Le Quang Dung:** Conceptualization, Resources, Supervision, Validation, Writing - review & editing. **Yin Fui Siau:** Investigation, Writing - review & editing. **Ahmad Nazila Bin Ali:** Investigation, Writing - review & editing. **Ibrahim Sunkanmi Saliu:** Investigation, Writing - review & editing. **Muhammad Amir Bin Fisol:** Investigation, Writing - review & editing. **Cristina Gonnelli:** Supervision, Validation, Writing - review & editing. **Farid Dahdouh-Guebas:** Conceptualization, Funding acquisition, Resources, Supervision, Validation, Writing - review & editing.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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