ZIBELINE INTERNATIONAL™ PUBLISHING

ISSN: 2773-6202 (Online)
CODEN: JTIOCC

Journal of Technology & Innovation (JTIN)

DOI: http://doi.org/10.26480/jtin.01.2022.01.05

RESEARCH ARTICLE

SEASONAL CHANGES OF TOTAL MERCURY IN A MANGROVE LAGOON ECOSYSTEM

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ARTICLE DETAILS

Article History:

Received 08 March 2022
Revised 05 May 2022
Accepted 12 May 2022
Available online 19 May 2022

ABSTRACT

In order to understand the variation of total mercury (THg) in a tropical mangrove lagoon ecosystem. THg concentration was seasonally analysed in the biota and surface sediments from Setiu lagoon, Terengganu, Malaysia. THg in sediments showed spatially the similar trend with higher concentrations in dry season compared to the rainy season, whereas most of the biota was found the opposite trend between seasons. Increased rainfall during the rainy season may be the main factor enhancing the bioavailability of THg to biota. Fishes are recorded the highest THg accumulation compared to other groups. However, the THg accumulation in carnivorous fishes suggests the need for a long-term THg monitoring programme in the mangrove lagoon.

KEYWORDS

sediment, biota, seasonal variations, bioaccumulation, impact, environment

1. INTRODUCTION

Mangrove systems have been recognized as one of the most productive ecosystems in the world, since they not only provide habitat to flora and fauna species, but also support offshore secondary production (Mitsch and Gosselink, 1993; Messina and Conner 1998; Ishikawa et al., 2003; Mumby, 2006; El-Regal and Ibrahim, 2014). However, mangroves also act as both major sink and trap the toxic metals such as mercury in aquatic ecosystem (Fitzgerald et al., 2007). Mercury (THg) is among persistent toxins of great concern, which can cause serious neurological disturbances to human and wildlife (Deocadiz et al., 1999; Wiener et al., 2003). Organic rich, anaerobic wetlands sediments provided ideal condition for sulphate-reducing bacteria to convert inorganic Hg to methylmercury (Hall et al., 2008). Hg is able to accumulate in biota and biomagnify through the marine food chain (Monteiro et al., 2010). Hg in fish is concerning as fish is the major protein source to human, and thus became major route of Hg exposure to human populations (Fitzgerald and Clarkson, 1991). However, the Hg accumulation in organisms may vary seasonally and spatially due to water and sediment quality, bioavailability of Hg to organisms, species variation, and the complexity of Hg react with the environment factor such as pH, dissolved organic carbon (DOC), salinity, and temperature (Kehrig et al., 2001; Laporte et al., 1997).

Setiu mangrove lagoon is a tropical natural wetland located in East Coast of Peninsular Malaysia with high biodiversity in both terrestrial and marine organisms (Amin and Hasan, 2003). The lagoon provides important fisheries to local people, however, the increasing of anthropogenic activities recently has affected water quality (Suratman and Latif, 2015). The elevated concentrations of heavy metals detected in sediment from a lake and river in the upper reach of the lagoon raised an environmental concern (Jamilah et al., 2015; Nik Fuad et al., 2003; Norhayati et al., 2006; Hanum et al., 2012). The objective of this study is to investigate seasonal changes of THg in Setiu mangrove lagoon.

2. MATERIALS AND METHODS

2.1 Sampling

The study area was located at lagoon of Setiu Wetlands in State Terengganu on The East Coast of Peninsular Malaysia. Samplings was carried out at wet (December – January) and dry (June - July) season in 2018. Two sites (STA1 and STA2) were designed for spatial sampling to determine the geographically THg distribution in the lagoon. Samples were collected including mangrove leaves, sediments, invertebrates, and fishes. Three types of mangrove leaves (green, senescent and decayed) were handpicked from the tree and the sediment surface. Sediments (1cm depth from the surface) were collected by shovel. Mollusca, such as bivalves and gastropods, were collected by hand during low tide at sampling sites. Oysters were collected from the prop roots of *Rhizophora apiculata*. However, crabs and fishes were collected using traps (40 cm length, 30 cm width and 25 cm height, the net mesh size is 2 cm). All samples collected were placed into labelled polyethylene bags and kept in ice box before transported back to laboratory. All samples were washed and stored at -20°C until further analysis.

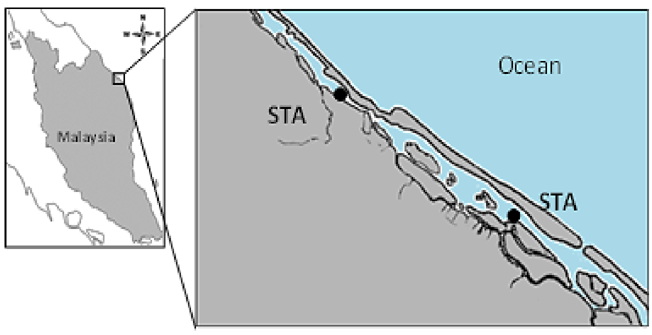

Figure 1: The sampling locations (●) from the Setiu Wetlands, in which, STA1 is sampling site 1 and STA2 is sampling site 2

2.2 Sample Preparation

In laboratory, biota samples were washed gently with distilled water followed by deionized water to remove unnecessary foreign materials. Samples were then identified to the lowest possible taxonomic level using

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Website: www.jtin.com.my

DOI: 10.26480/jtin.01.2022.01.05

Cite The Article: Siau Yin Fui, Nguyen Thanh Luan, Nguyen Phan Thao, Dung Quang Leb Fukad (2022). Seasonal Changes of Total Mercury in A Mangrove Lagoon Ecosystem. *Journal of Technology & Innovation, 2(1): 01-05.*

taxonomy books of the Invertebrates of Setiu Wetlands and Fishes of Terengganu (Faridah et al., 2017; Matsunuma et al., 2011). Length and weight of fish samples were measured and recorded. Fish samples were classified into three groups, carnivore, omnivore and herbivore. Fish were then dissected, and white dorsal muscle tissues were taken for further analysis. Muscle tissues of gastropods, bivalves and crustaceans were removed from their shells and chelae. All samples were dried at 60°C until constant weight was obtained (12 hours). Wet and dry weight of the samples were recorded, and the dried samples were ground into powder form using mortar and pestle.

2.3 Mercury Analysis

The THg concentration of powdered samples were directly measured using cold vapour atomic absorption spectrometer (Model MA-3000, Nippon Instruments Corp., Japan) with detection limit of 0.0002 mg/kg. The quality controls for the total Hg measurements include blanks, replicates, Hg standard (L-cysteine) solutions. The precision and accuracy of the method was determined using standard reference material NIST-SRM2976 (National Institute of Standards and Technology, USA) in all sample batches. 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 concentration (mg/kg dry weight) in each fish sample was generated from 0-, 0.5-, 1.0- and 2.0-ppm standards. The recovery of THg was ranged from 90 – 105%.

2.4 Statistical Analyses

Results are expressed in mean ± SD. Shapiro-Wilk test were performed to test the normality of the data. Since most of the variables were normally distributed, parametric tests were performed for statistical analysis. ANOVA test and stepwise regression were used to test the significant differences of THg concentrations among taxa groups, and also the influence of water parameter, trophic position and body length to THg concentration in samples. Independent-samples T test were performed to reveal significant differences of samples THg concentration between seasons and sites.

3. Result and Discussion

THg concentration in biota and surface sediments are expressed in dry weight basis as shown in Table 1. Of the samples measured, THg concentration in both seasons followed a decreasing trend of: fish > crustaceans > bivalves > gastropods > sediments > mangrove leaves. THg concentration in fish were significantly higher than all the other taxa group of samples ($p<0.05$). However, there were no significant differences among crustaceans, bivalves, and gastropods in THg concentrations. THg concentration of fishes, bivalves, gastropods and mangrove leaves collected in wet season were significantly higher than those in dry season, whereas sediments shows higher THg concentration in dry season compared to wet season ($p<0.05$). Crustaceans did not present any significant differences between seasons.

Table 1: Thg Concentration in Samples Collected in Setiu Wetlands

Season	Sample	N	STA1		N	STA2	
			Mean ± SD (µg/kg)	Range (µg/kg)		Mean ± SD (µg/kg)	Range (µg/kg)
Wet Season (January 2018)	Fish	44	422.67±381.61	42.88-1656.80	25	535.34±393.75	122.37-1499.80
	Crustaceans	8	170.69±135.27	39.42-424.43	5	80.10±36.23	44.29-128.74
	Bivalves	34	234.12±140.17	40.00-507.38	14	178.19±136.88	63.59-451.71
	Gastropods	40	111.42±38.30	58.88-193.19	17	197.36±98.18	104.51-429.90
	Mangrove Leaves	9	48.41±9.20	36.97-69.72	9	20.46±11.67	6.04-40.67
	Sediment	5	51.57±3.34	47.70-56.75	5	72.73±3.09	68.18-76.68
Dry Season (July 2018)	Fish	76	195.98±149.38	19.94-796.29	64	227.57±123.35	55.033-567.98
	Crustaceans	13	108.92±59.96	46.16-257.63	11	228.58±205.99	32.37-568.23
	Bivalves	18	73.02±44.14	43.49-233.82	19	103.11±40.63	27.38-184.22
	Gastropods	25	97.70±48.37	33.62-183.35	26	130.61±45.61	62.12-206.81
	Mangrove Leaves	10	18.45±5.93	10.03-26.85	11	24.87±10.27	9.74-35.62
	Sediment	6	71.31±8.46	62.45-81.04	6	76.93±8.84	64.37-88.95

Overall, THg concentration in sediments ranged within 47.70 to 88.95µg/kg. There was significant higher THg concentration in sediments during dry season compared to wet season ($p<0.05$). Similar trend of sediments Hg was shown in STA2 however the seasonal variation was not significant (Figure 2). Lower concentration of THg in sediment in wet season compared to dry season could be due to the disturbance of surface sediment by the strong water turbulence flow and released of THg from sediment into water column during rainy season. Sedimentation is strongly depending on the particle size and current strength. Small sediment particle size and strong current flow are tended to slower the sedimentation rate (Nybakken, 2005).

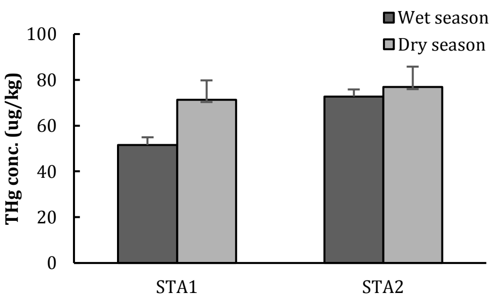

Figure 2: THg Concentrations in Sediments from Lagoon Setiu Wetlands

THg concentration in mangrove leaves (*Rhizophora apiculata*) varied from 6.04 to 69.72µg/kg. In STA1, THg concentration in mangrove leaves showed higher value in wet season compared to dry season ($p<0.05$), while there is no significant difference between seasons in STA2. In all sites and seasons, THg concentration variation across the leaf senescence stages was observed, where higher THg concentration was found in decayed and yellow leaves than in green leaves ($p=0.003$). Increasing of THg concentration in mangrove leaves during their lifespan was observed, where decayed and yellow leaves contain higher THg than green leaves (Figure 3). Previous study reported similar increasing trend of THg with aging of mangrove leaves (Ding et al., 2011). Elimination of heavy metal through direct excretion or leaf fall had been suggested as detoxification mechanism of plants, where heavy metals are accumulated in senescent leaves and released back into environment when leaves become detritus (Dahmani-Muller et al., 2000; Windom et al., 1976). THg exchange between decayed leaves and the environment also contributed to the changes of THg in plant litter, where THg is lost through decomposition, or increased through absorbing of Hg from water body in wetlands (Zhuang and Lin, 1992). Release of Hg through plant litter therefore become a source of Hg to the organisms.

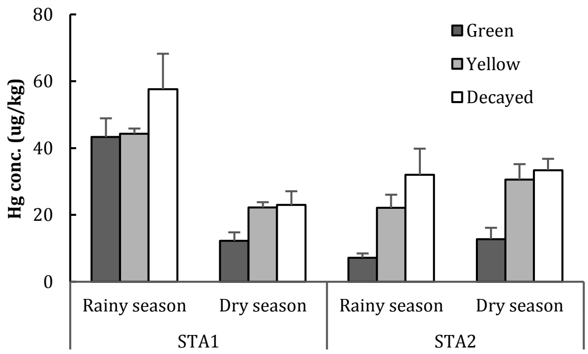

Figure 3: THg Concentrations of Mangrove Leaves from Lagoon Setiu Wetlands

Mean THg concentration of invertebrate varied from 33.62 - 429.90μg/kg in gastropods, 27.38 - 507.38μg/kg in bivalves, and 32.37 - 568.23μg/kg in crustaceans in dry weight basis. Unlike sediments, both gastropods and bivalves showed inverse trend of THg accumulation, where THg concentration in wet season is significantly higher than in dry season ($p<0.05$) in both sampling sites (Figure 4, Figure 5). In both season, gastropods in STA2 showed higher THg concentration compared to STA1 ($p<0.05$). THg accumulation variation in biota depend on several factors such as metal bioavailability, hydrodynamics of environment, seasonal variation, size, sex, growth rate, feeding behaviour and detoxifying mechanism of biota (Boyden and Philips, 1981; Ruelas-Inzunza and Páez-Osuna, 2000).

Benthic organism such as gastropods and bivalves are frequently used as bio-indicator of metal pollution in aquatic ecosystem. Bivalves are filter feeders which generally feed on suspended matter in that available in water column. Previous study also found the ability of bivalves to absorb Hg from water and food (Kehrig et al., 2006). In present study, bivalves in both sampling sites has shown higher THg concentration in wet season, which is an opposite trend with the sediment THg concentration. This suggested the result of THg released from sediment during wet season has increased the chance of THg uptake by bivalves from the overlying water column. Besides that, the runoff of THg waste from upland activities into water column with the heavy rain may also contribute to the elevation of THg concentration in water column. However, THg concentration in bivalves varied greatly among species (oyster > marsh clams > asian hard clams > blood cockles).

This variation suggested the difference on food preference and detoxifying mechanism of bivalves. Compare of THg accumulation in five species of marine bivalves had found that oysters (*Saccostrea cucullata*) had the lowest excretion rate of THg and MeHg compared to other bivalves species (Pan and Wang, 2011). Oysters *Crassostrea rhizophorae* was reported with lower THg concentration than mussel *Perna perna* (Kehrig et al., 2006). The study suggested the variation may due to the food preference, where mussels feed on higher particle size (organic detritus, silt and nanozooplankton) compared to oyster (phytoplankton). Blood cockles presented lower THg concentration compared to oysters and clams in this study. Cockles are usually found in relatively saline area, and in the sediments with higher sand proportion. Sediments with larger grain size are less likely to accumulate THg and hence, blood cockles did not receive much THg from sediments as compared to bivalves in mangrove area (Bryan et al., 1985).

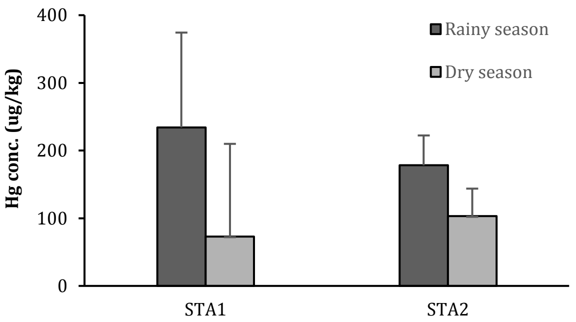

Figure 4: THg Concentrations of Bivalves from Lagoon Setiu Wetlands

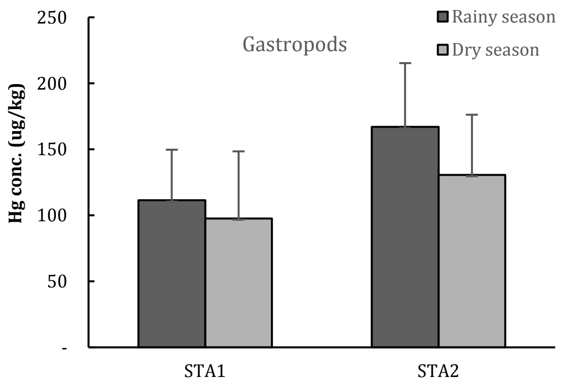

Figure 5: THg Concentrations of Gastropods from Lagoon Setiu Wetlands

Some studies has demonstrated close association of gastropods with mangrove vegetation. Potimididae was one of the gastropods families which strongly associated with mangrove, and most of the species in genus *Littoraria* can only be found on mangrove trees. *Cassidula* and *Ellobium* are found to be restricted to mangroves (Reid et al., 2008; Reid, 2011; Ozawa et al., 2015). Abundance of gastropods increased with the increases of mangrove vegetation age, which related to higher C-organic content and litters fall found in old-growth mangrove forest (Irma and Sofyatuddin, 2012). In this study, gastropods has been collected from both mangrove and seagrass area. Deposit-feeding gastropods species collected from seagrass area (*Faunus ater, Clithon oualaniensis*) shown significant higher THg concentration in wet season compared to those collected in dry season, whereas mangrove-associated gastropods did not show any seasonal variation in THg concentration. This may due to distinct feeding behavior and habitat differences among species. *Faunus ater* and *Clithon oualaniensis* inhabit on mud flat, feed on detritus, algae and diatoms on seagrass (Rintelen 2011; Ng et al., 2011; CSIRO, 2007). Long-term exposure of deposit-feeding gastropods to water column and sediment has caused the THg concentration in biota was highly affected by the environment changes. Increase of THg input and release of THg from sediment into water column during rainy season probably led to increase of THg uptake rate in gastropods.

On the other hand, sesarmid crab did not present any significant seasonal and spatial change. While portunid crabs (*Thalamita crenata, Scylla olivera, Portunus pelagicus*) in STA2 contained higher THg concentration than STA1 (Figure 6). THg concentration in crustacean samples varied greatly may due to distinct feeding behaviour and habitat. Sesarmid crabs are found to be herbivores which majorly feed on mangrove litter and mangrove-derived carbon sources (Poovachiranon and Tantichodok, 1991; Ravichandran et al., 2006; Wu and Shin, 1997). Thus, sesarmid crabs are less likely to accumulate THg as much as carnivorous crab. As the predator in higher trophic level, portunid crabs (*Scylla olivacea, Thalamita crenata, Portunus pelagicus*) which prey on various food item such as bivalves and slow-moving crustaceans tend to receive more THg from the preys (Ravichandran et al., 2006; Wu and Shin, 1997; Kunsook et al., 2014). Portunid crabs in STA2 had higher THg than those in STA1, and this may related to the averagely higher THg in food sources from STA2.

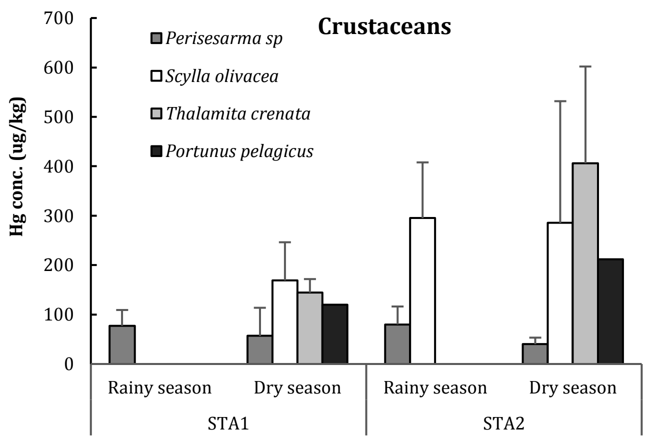

Figure 6. THg Concentrations of Crustaceans from Lagoon Setiu Wetlands

Distribution of THg in fish varied greatly, which from 19.94-1656.80μg/kg. Fishes collected during wet season contained higher THg concentration than those collected during dry season ($p<0.05$). This may be due to variation of fish composition between wet and dry season. Distributions of fish are related to the physiological tolerances of species to the salinity fluctuations of the environment for enhancement of the survival, growth and body condition (Blaber, 1997; Lin and Shao, 1999). Relationship among THg concentration with feeding behaviour and trophic level was observed. THg concentration in carnivorous and omnivorous fish were significantly higher than herbivores (Figure 7, $p<0.05$).

Accumulation of THg in fish are known to be species-specific and this is found to be relevant the different feeding behaviour, size, trophic level, life span, and detoxifying mechanism of different species (Monteiro et al., 1991; Bank et al., 2007; Evans et al., 2005; Ahmand et al., 2015). Result of this study was found to be consistent with other studies where higher concentration was found in carnivorous fish with higher trophic level (Evans et al., 2005; Ahmand et al., 2015; Burger and Gochfeld 2011; Kasper et al., 2009). Previous study also found out that carnivorous fish contained higher ratio of organic mercury compared to inorganic Hg (Kasper et al., 2009). Furthermore, fish in high trophic level may exposed to varieties of prey type and larger prey that highly contaminated with Hg. Thus, fish in higher trophic level tend to accumulate more Hg in their body.

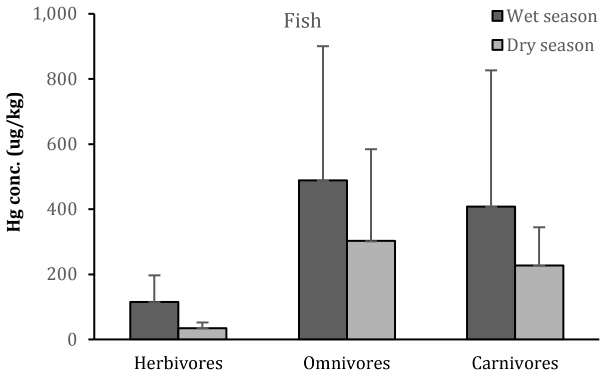

Figure 7: THg Concentrations of Crustaceans From Lagoon Setiu Wetlands

4. Conclusion

Changes of abiotic environmental factors across seasons are likely to influence the trends of THg accumulation. The elevated THg accumulation in some fish species, particularly carnivores, suggests the need for a long-term THg monitoring programme in the mangrove lagoon.

Acknowledgement

The authors would like to express their gratitude to staffs in Universiti Malaysia Terengganu for their assistance and guidance in field and lab session. The present work has been done with the support from Ministry of Higher Education Malaysia, under Fundamental Research Grant Scheme (FRGS) project number FGS/1/2016/WAB09/UMT/02/5 (59425).

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