論文内容の要旨

論文題目

MOBILITY OF TRACE ELEMENTS IN THE MANGROVE ECOSYSTEM OF PENINSULAR MALAYSIA

（マレーシア半島のマングローブ生態系における金属類の挙動）

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Abstract

To understand the mobility of trace elements from the environment and transfer across trophic levels in mangrove ecosystem, several associated studies have been conducted including study of food preference of the giant mudskipper (*Periophthalmodon schlosseri*), assessment of trace elements in sediments, mudskippers and its food items, and designing an ecological model of trace elements mobility in mangrove ecosystem. Several methods have been adopted including stomach content and stable isotopes analyses, and assessment of trace elements in sediments and biota tissues. Results showed trace elements level in sediments of Peninsular Malaysia were range from “unpolluted” to “very severely polluted”, depending on activities occurring around the sampling area. Stomach content and stable isotopes analyses showed feeding preference of the mudskipper is depended on sexes and life stages. Bioaccumulations of trace elements in organs of the mudskipper were closely related with the functions and existing physiological activities. Trace elements were found low in lower trophic level and increase with increasing trophic level. Combination of stable isotopes analysis and trace elements concentrations in tissue samples showed biomagnification of certain trace elements occurred in the mangrove ecosystem.

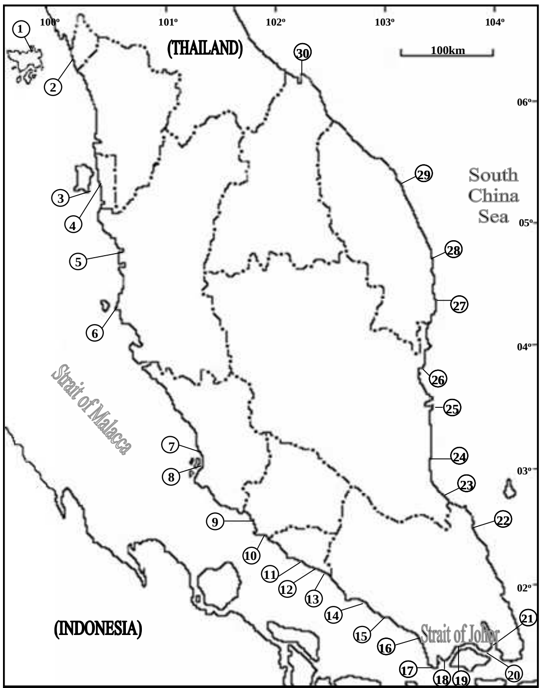
Introduction

In present days, the field of ecotoxicology has become an important subject since various kinds of pollutants that are reported or potentially threats the environment and biota, including trace elements, dioxin, PCB, PAHs etc. Locally in Peninsular Malaysia, a deliberate but increasing understanding and paradigm shift on environmental issues among professionals and public communities has offered a new hope on resolving existing problems. Current practice on assessing and monitoring chemical contamination still apply conventional approaches in which studying effects of chemicals on separate organisms and hypothesize their effects on consumers at the upper level along the food chain. Present study used a novel approach to study the mobility of trace elements in the mangrove ecosystem. In order to justify the main objective, a few objectives were set: (1) assess the background level of trace elements in the surface sediments; (2) study the food preference of the sentinel species in mangrove area, the giant mudskipper

(*Periophthalmodon schlosseri*); (3) determine bioaccumulation of trace elements in *P. schlosseri* and its' food items: (4) design a conceptual model for mobility of trace elements in the mangrove ecosystem

Materials and Method

Surface sediments were collected from intertidal areas around Peninsular Malaysia (Fig. 1). *P. schlosseri* and its' food items were collected from the mangrove area of Puloh River (Site # 7 on the map). All samples were stored at cooled temperature (-20^{o}C) prior further analytical procedures. Assessments of trace elements were conducted according established methods including the aqua-regia method (ISO11466) for sediments, and microwave-assisted digestion method (Yang et al. 2004) for biota. Concentrations of trace elements were determined using an ICP-MS. The food preference of *P. schlosseri* was determined using the stomach content- (Hyslop 1980; Cortés 1997) and stable isotopes- (Nakamura et al. 2008) analyses. Pollution status in respective sampling station was determined by comparing with the Interim Sediment Quality Guidelines (ISQGs) of ANZECC/ARMCANZ and Hong Kong, and calculations of the enrichment factors (*EF*) and the geoaccumulation index (I_{geo}). Accuracy of analytical methods were determined using certified reference materials (trace elements: SRM 1646a, DORM-2 and SRM 1577b; stable isotopes: atmospheric nitrogen (N_2) and Vienna- Pee Dee Belemnite (V-PDB)) with acceptable recovery values.



1.	Tanjung Rhu (LTR)	16.	Pontian Kecil (JPK)
2.	Kuala Perlis (RKP)	17.	Tanjung Piai (JTPi)
3.	Bayan Lepas (PBL)	18.	Tanjung Pelepas (JTPe)
4.	Kuala Perai (PKP)	19.	Pantai Lido (JPL)
5.	Kuala Sepetang (AKS)	20.	Kampung Pasir Putih (JPP)
6.	Lumut Port (ALP)	21.	Kong Kong (JKK)
7.	Puloh River (BSP)	22.	Mersing Jetty (JMJ)
8.	Tanjung Harapan (BTH)	23.	Kuala Rompin (CRR)
9.	Port Dickson (NPD)	24.	Nenasi River (CNR)
10.	Pasir Panjang (NPP)	25.	Kuala Pahang (CKP)
11.	Melaka River (MMR)	26.	Kuantan River (CKR)
12.	Serkam River (MSeR)	27.	Bukit Kuang (TBK)
13.	Sebatu River (MSR)	28.	Kuala Dungun (TKD)
14.	Parit Jawa (JPJ)	29.	Kuala Terengganu (TKT)
15.	Minyak Beku (JMB)	30.	Kuala Besar (DKB)

Fig. 1: Sampling location of sediments around Peninsular Malaysia

Table 1: Comparison of trace elements of present study with other studies around the world

Element	Peninsular Malaysia, Malaysia (present study)	San Francisco Bay, USA (Hwang et al. 2008)	North Sulawesi, Indonesia (Edinger et al. 2008)	Mangrove area, Singapore (Cuong et al. 2005)	Szczecin Lagoon, Poland (Glasby et al. 2004)	Florida Bay, USA (Caccia et al. 2003)	Strait of Johor, Malaysia (Wood et al. 1997)
V	12-276	90-122	26-182	-	34-59	34-411	23-119
Cr	11-207	135-241	8-52	9-40	-	60-347	13-66
Co	1-15	17-25.4	3-21	-	7.1-17	0.6-10	2-8
Ni	2-36	110-145	4-40	4-16	18-56	5-54	21-34
Cu	2-151	101-541	10-63	1-46	20-103	7-32	11-93
Zn	24-609	280-1430	34-932	11-134	256-1310	10-48	54-334
As	0.1-312	21-164	5-275	-	-	-	7-39
Ag	n.d.-27	0.7-1.73	-	-	-	-	-
Cd	n.d.-1.1	0.7-7.75	-	n.d.-0.4	1-6	-	0.1-0.3
Pb	8-93	218-750	3-12	7-37	42-167	3-16	19-160
U	0.7-20	-	-	-	2-4	-	3-7

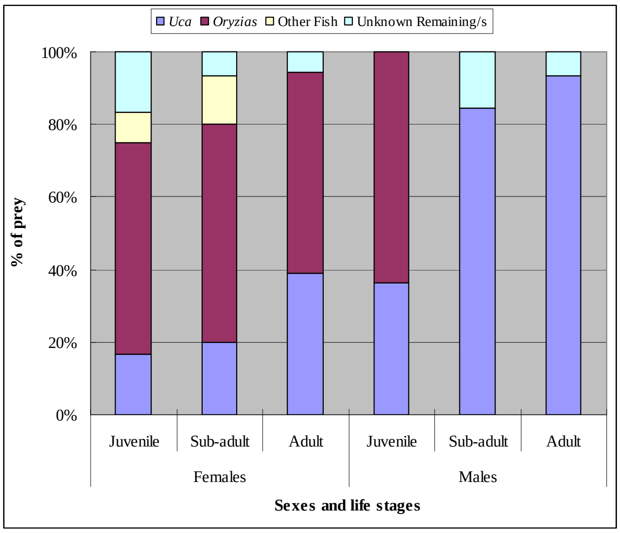
Results and Discussion

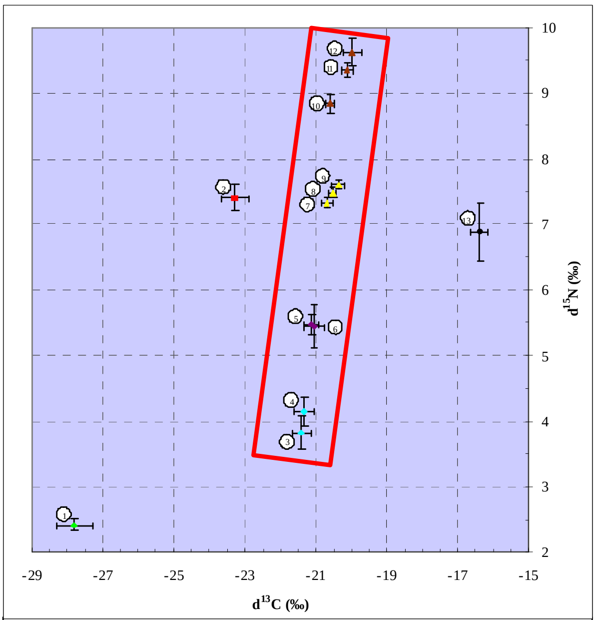
1) Trace elements in sediment of Peninsular Malaysia

In general, concentration of trace elements in surface sediments can be arranged as follows: Fe > Mn > Zn > V > As > Cr > Pb > Cu > Ni > Co > U > Mo > Ag > Cd. Concentrations of trace elements obtained in present study were within the range of other studies conducted around the world, except for V, As and U (Table 1). Data screening using a few simple calculation showed only certain locations around Peninsular Malaysia had mean concentration above ISQG-high and/or ISQG-low, thus raise a possibility that some biota species have been affected by these elements in the involved areas. After calculation using the *EF* and I_{geo}, trace elements in sediments were ranged between "no enrichment" and "extremely sever enrichment", and "unpolluted" and "very strongly polluted", respectively. Developed areas with high density of manufacturing industries, agricultural based industries (oil palm, rubber, etc.), husbandry and poultry farms, transportations, landfills, domestic wastes industrial, population and sea vessels were areas strongly related with high contamination of trace elements (e.g. Pasir Gudang, Lumut, Perai, Klang etc.). There for, we concluded that contamination of trace elements were localized in vicinity of nearby sources.

2) Food preference of *P. schlosseri*

As for the food preference of *P. schlosseri*, combination of stomach content analysis and stable isotope analysis was found to be perfect tools to investigate this matter (Fig. 2). Although the stomach content analysis revealed that there were four groups of food items for *P. schlosseri* (*Uca* sp., *Oryzias* sp., juveniles of other fish species, and unknown remainings), later the stable isotope analysis managed to exclude uncommon food items. The main food items were verified as *Uca* sp. and *Oryzias* sp. only. The unknown remainings were found to be either digested or partly digested of these main food items. Selection of food





Remark:
1 - *Rhizophora* sp. leaf
2 - Juvenile of other fish species
3 - *Oryzias* sp.
4 - Unknown remainings (female *P. schlosseri*)
5 - Unknown remainings (male *P. schlosseri*)
6 - Small *Uca* sp. (<25mm)
7 - Juvenile female *P. schlosseri*
8 - Sub-adult female *P. schlosseri*
9 - Adult female *P. schlosseri*
10 - Juvenile male *P. schlosseri*
11 - Sub-adult male *P. schlosseri*
12 - Adult male *P. schlosseri*
13 - Big Uca sp. (>25mm)

Fig. 2: Percentage of prey items according life stages of female and male *P. schlosseri* (a), and δ^{13}C and δ^{15}N ratios of biota (b) from mangrove area of Puloh River

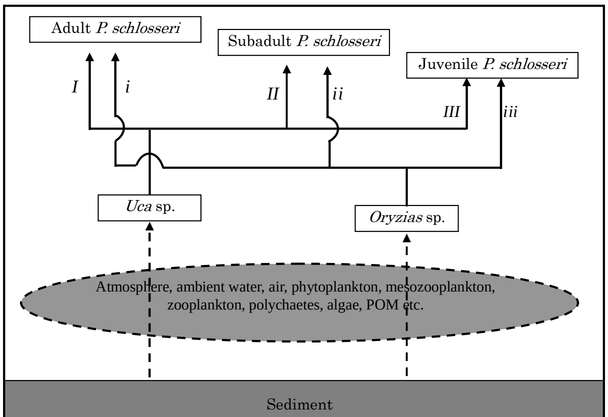
items was also influence by sexes and maturity stages, possibly due to different behavior and living strategy.

3) Trace elements in *P. schlosseri* and its' food items

The next stage was to assess trace elements concentration in *P. schlosseri* and its' food items. Bioaccumulation trend of selected trace elements in *P. schlosseri* can be arranged as follows: Fe > Zn > Mn > Cu > Cr > As > Ni > Pb > Cd. There was no significant difference in bioaccumulation of most trace elements among different sexes, except for Ni, Cu and As. However, bioaccumulation of trace elements was found significantly different among different maturity stages. Distribution of these elements was varied, depending on functions and metabolic activities occurring in each organ. In general, the most of highly bioaccumulated elements were found in detoxifying organs (spleen, liver and kidney) and those having calcified structures (bone and gills). In some occasions, other organs (i.e. brain, eye and gonad) were found bioaccumulated significantly high concentration certain elements due to adaptation and specific purposes. For the food items of *P. schlosseri*, *Uca* sp. was found bioaccumulated with higher concentration trace elements than *Oryzias* sp., except for Ni, Cu and As. In general, Cu and Zn was found occurring at higher concentration in invertebrates than their predator at the upper trophic level(s) since they are known to bound at metal-rich granules and sequestered as a detoxified forms, prior excretion out of their body.

4) Mobility of trace elements in mangrove area

Finally is about the mobility of trace elements in the mangrove ecosystem in relation with stable isotopes (δ^{13}C and δ^{15}N) ratios. Concentration of trace elements in each biota was correlated with the stable isotope ratios. Correlation analysis was used in present study to verify biomagnification process occur at the upper trophic level in the mangrove ecosystem of Puloh River. All non-essential trace elements (Ni, As, Cd and Pb) certain essential trace elements (Cr, Mn, Fe and Zn) were found biomagnified along the food chain. Cu was not biomagnified. A conceptual ecological model for the mobility of trace elements in the mangrove ecosystem was design to give clear illustration of transporting process of trace elements across trophic levels (Fig. 3). Biomagnification factors were calculated for respective trace elements and the results showed the factors increased with the increase



Label	$\Delta\delta^{15}$N	Biomagnification Factor						
		Cr	Mn	Fe	Ni	As	Cd	Pb
I	1.40x	9x	5x	2.9x	6x	2.2x	6x	4x
II	1.38x	7x	4x	2.3x	5x	1.7x	4x	3x
III	1.35x	5x	3x	1.8x	4x	1.3x	3x	2x
i	1.99x	18x	7x	4x	2.4x	7x	10x	58x
ii	1.96x	14x	6x	3x	1.9x	5x	8x	45x
iii	1.92x	11x	4x	2x	1.5x	4x	6x	35x

Fig. 3: A conceptual ecological model of the mobility of trace elements in mangrove ecosystem

of trophic level.

Conclusion

In conclusion, present study identified trace elements contaminations in sediments around Peninsular Malaysia were localized in vicinity of their sources. Bioaccumulation of trace elements in *P. schlosseri* was found highly bioaccumulated in detoxifying organs and calcified structures. Combination of stomach content and stable isotope analyses was found as ideal combination to identify the food preference of *P. schlosseri* from Puloh River, which comprised only *Uca* sp. and *Oryzias* sp. Biomagnifications of toxic elements (Ni, As, Cd, Pb) and some essential trace elements (Cr, Mn, Fe) along food chain were proven occurred in the mangrove ecosystem.

Recommendation

Further extensive and comprehensive studies involving assessment of stable isotope ratios and trace elements concentration of organisms at the lower trophic levels will be conducted to improve the conceptual models of trace elements mobility in mangrove ecosystem.