

Correlations between Geo-Chemical Speciation of Heavy Metals (Cu, Zn, Pb, Cd and Ni) in Surface Sediments and Their Concentrations in Giant Mudskipper (*Periophthalmodon schlosseri*) Collected from the West Coast of Peninsular Malaysia

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Abstract

Surface sediments and giant mudskipper (*Periophthalmodon schlosseri*) were collected in August and September 2008 and in March and June 2010 from six sampling sites in the west coast of Peninsular Malaysia to assess heavy metals accumulation in the giant mudskipper. Sequential extraction technique was used to fractionate the sediments into four different geo-chemical fractions; easily, freely or leachable and exchangeable (EFLE), acid reducible, oxidizable organic and resistant fractions. Heavy metals concentrations (Cu, Zn, Pb, Cd and Ni) in the surface sediments and giant mudskipper were determined by using air acetylene flame atomic absorption spectrophotometer (AAS) Perkin Elmer Analyst 800. The results of Pearson's correlation analyses showed that metal concentrations in the tissues of *P. schlosseri* were significantly correlated ($p < 0.01$ or $p < 0.05$), correlations were observed between Cu in *P. schlosseri* and Cu in the sediment (oxidisable-organic, resistant and total Cu), Zn in *P. schlosseri* and Zn in the sediment (EFLE and total Zn), Pb in *P. schlosseri* and Pb in the sediment (with all the four fractions of Pb), Cd and Ni in *P. schlosseri* and Cd and Ni in the sediment (with all fractions of Cd and Ni except acid-reducible Cd and Ni) which might suggest the use of *P. schlosseri* as a biomonitoring agent for heavy metals pollution in the west coast of Peninsular Malaysia.

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Keywords

Surface Sediments, Heavy Metals, Geo-Chemical Speciation, *Periophthalmodon schlosseri*, West Coast of Peninsular Malaysia

1. Introduction

Rapid socio-economic growth and industrialization in the coastal area has led to the discharged of heavy metals into aquatic environments and their accumulation in sediments and living organisms to a toxic level. The pollution of aquatic systems had become a major concern worldwide [1] (Abdel-Baki, Dkhil, and Al-Quraishy, 2013). Sediment-associated metals pose a direct risk to detrital and deposit-feeding benthic organisms, and may also represent long-term sources of contamination to higher trophic levels [2]-[5]. The bioavailability and subsequent toxicity of the metals are dependent upon various kinds and amounts of the metals bound to the sediment matrices (metal speciation) [6]. It had been found that the speciation of a metal, rather than its total concentration, is the key to understand its effect on the biota [7] (Allen and Hansen, 1996), as well as its biogeochemical transformation and ultimate fate [8] (Billon *et al.*, 2002). Increased discharge of both essential and non-essential metals into natural aquatic ecosystems could expose aquatic organisms to unnaturally high levels of these metals [9] (Van Dyk *et al.*, 2007), thereby causing adverse effects on aquatic ecosystem. If these metals were taken up excessively, they might be detrimental to aquatic organisms [10] (Chourpagar & Kulkarni, 2011). Many biomonitoring agents for heavy metals were proposed in the west coast of Peninsular Malaysia but the use of mudskippers in the biomonitoring of Malaysian coastal environments had not received much attention like other biota. Mudskippers were peculiar-looking amphibious fishes that were characteristic for mangrove forests and mudflats [11] (Kruitwagen, 2007); that were able to accumulate metals. Considering the continuous input of heavy metals in coastal environment (mudflat areas) and function of mudskipper as key specie in intertidal mudflat area, correlation between concentrations of heavy metals in different sediment fractions and tissues of giant mudskipper *P. schlosseri* levels could give better information on the bioavailable metals in sediments. The objectives of the present study was to assess the bioaccumulation of Cu, Zn, Pb, Cd and Ni in giant mudskipper *P. schlosseri* and their correlations with different geochemical fractions of sediments.

2. Materials and Methods

Samplings were conducted in August 2008 to June 2010. A total of six sampling sites were selected for surface sediments and giant mudskipper *P. schlosseri* in the west coast of Peninsular Malaysia (Figure 1).

The coordinates of sampling sites were recorded with Global positioning system (Garmin OREGON 450T 850 MB waterproof GPS). Description of each sampling site was given in Table 1. Top 3 - 5 cm surface sediments [12] [13] (Ismail, 1993; Zulkifli *et al.*, 2010) were collected in triplicates from three different points within a certain area (approximately 1 meter radius) from each sampling site using plastic scoop and placed in separate labelled polyethylene plastic bags. Each sediment sample was instantly placed in ice and transported to the laboratory until further analysis.

Sediment samples were dried in the laboratory using an air-circulating oven to a constant dry weight at 80°C. The dried sediment samples were crushed to powder by using a porcelain mortar and pestle then sieved vigorously to produce homogeneity [12] (Ismail, 1993), through a 63 µm stainless steel aperture sieve. Geochemical fractions of Cu, Zn, Pb, Cd and Ni in the sediments were determined by modified sequential extraction technique (SET) as described by [14] Badri and Aston (1983) and [15] Tessier and Campbell (1987).

The four-step extraction procedure of sediment handling and analysis employed using SET was described as: 1) easily, freely or leachable and exchangeable (EFLE) (Fraction 1); 2) acid reducible (Fraction 2); 3) oxidisable organic (Fraction 3) and; 4) resistant fractions (Fraction 4). The mathematical summation of the first three types of fractions constitutes the “non-resistant phase which is closely related to anthropogenic inputs” [14] (Badri and Aston, 1983).

The residue used for each fraction was weighed before the next fractionation was carried out. The residue was rinsed with 20 mL DDW, filtered through a Whatman® No. 1 filter paper, and the filtrate stored until metal determination. For each fraction of the sequential extraction procedure, a blank was employed using the same

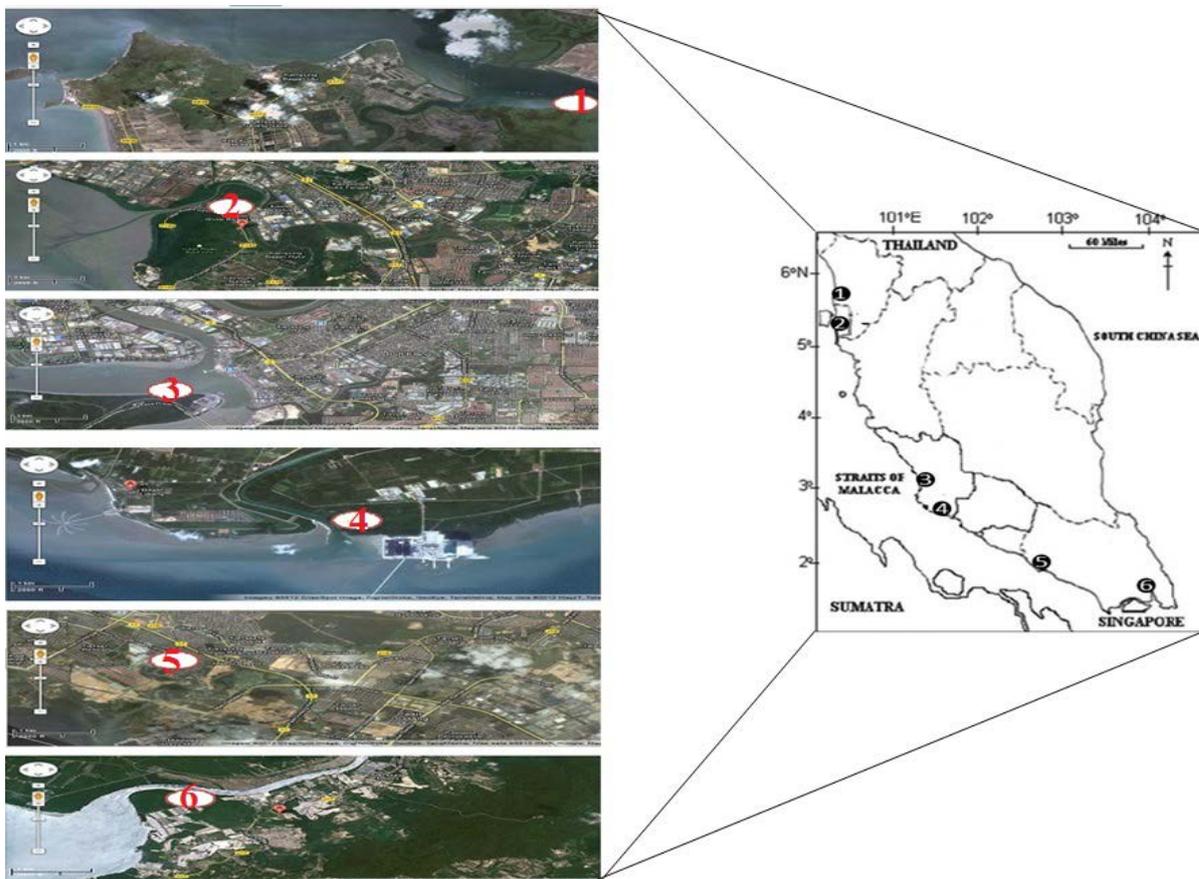


Figure 1. Map of west coast of Peninsular Malaysia showing six sampling sites (1, 2, 3, 4, 5 and 6).

Table 1. Description of sampling sites in the west coast of Peninsular Malaysia.

No.	Sampling site	Description of sampling sites	Coordinates	Site description
1	Sengantang Garam, Kedah		05°39.552'N 100°23.983'E	Jetty, aqua cultural area and paddy field
2	Kuala Juru, Penang		05°19.683'N 100°22.949'E	Industrial area
3	Sungai Puluh, Klang		03°04.786'N 101°23.903'E	Jetty receiving domestic wastes and industrial area
4	Bagan Lalang, Selangor		02°36.669'N 101°41.100'E	Recreational and agricultural areas
5	Minyak Beku, Johor		01°47.746'N 102°53.395'E	Jetty receiving domestic wastes and shipping activities
6	Sungai Tiga, Johor		01°25.841'N 104°00.281'E	Jetty, agricultural and oil plantation

procedure to ensure that samples and chemicals used were free of contaminations.

Fish were collected using trap net at almost the same locations where the sediments were collected and placed in a plastic aquarium containing some sediment and water. The fish samples were brought to the laboratory; stomach and intestines were emptied and dissected immediately or put in labelled plastic bags and kept in deep freeze at -20°C until further analysis. In the laboratory, fish samples were removed from the refrigerator and plastic bags, rinsed with DDW then thawed at room temperature. About 10 *P. schlosseri* from each sampling sites with the recorded body length (mm) and weight (g) were dissected on clean plastic material using stainless steel kits and glass equipment. The dissected parts were pooled into eight different parts namely; scales, muscle,

bone, gills, operculum, intestine, liver and cartilage. All the eight parts were dried at 80°C according to [16] (Mucha *et al.*, 2003) until a constant dry weight.

Sample of each dried part was weighed separately (0.5 - 1.0 g) in triplicate and placed in digestion tubes. To each digestion tube 10 ml concentrated nitric acid (Anala R grade, BDH 69%) was added and placed in a hot block digester unit at 40°C for 1 hour. The temperature was then increased to 140°C for at least three hours [12] (Ismail, 1993). The digested samples were diluted to 40mL with double distilled water (DDW). The samples were then filtered through filter papers into pill box and the filtrate was stored until metal determination.

The filtrates obtained from sediments and biological samples were determined for Cu, Zn, Pb, Cd and Ni by using an air-acetylene flame atomic absorption spectrophotometer (AAS) Perkin Elmer Analyst 800. Standard solutions were prepared from 1000 mg/L stock solution of each metal (BDH SpectrosoL).

The wavelengths for each metal were 324.8, 213.9, 283.3, 228.8 and 232.0 nm for Cu, Zn, Pb, Cd and Ni respectively. The data were presented in µg/g dry weight. Multiple-level calibration standards were analysed to generate calibration curves against which sample concentrations were calculated.

During the period of AAS metal analysis, a quality control sample was routinely included for every 5 - 10 samples. Procedural blanks and quality control samples made from standard solutions for Cu, Zn, Pb, Cd and Ni were analyzed after every 5 - 10 samples to ensure the sensitivity and recovery of the instrument used.

The procedures of quality assurance (QA) and quality control (QC) were employed to ensure the validity of the analytical data [17] (Ismail *et al.*, 2004). All plastics and glassware used were washed with detergent, Deacon 90, rinsed with double-distilled water and soaked in 10% HNO₃ for at least 24 hrs, then rinsed with double-distilled water and allowed to dry at room temperature. The QA and QC were controlled by procedural blanks, sample replicates and certified reference material (CRM).

The quality of the method was checked with a certified reference material (CRM) for soil from International Atomic Energy Agency (IAEA), Soil-5, Vienna; Austria and Dogfish liver DOLT-3 from National Research Council Canada (NRCC) were analyzed. These were checked to accuracy of the digestion method with the certified values supplied by the IAEA and NRCC. To ensure the sensitivity of the Atomic Absorption Spectrophotometer (AAS) and generate calibration curves against which sample concentrations were calculated. The results of similar digested samples analyzed for Cu, Zn, Pb, Cd, and Ni by the flame AAS Perkin Elmer. An analyst 800 showed acceptable recoveries of the metals; about 94% - 107% for soil and 87% - 110% for dogfish liver. All statistical analyses of data were carried out using SPSS statistical package programs version 17.

Data were tested for the basic assumptions of normality and homogeneity of variance in exploratory data analysis in SPSS 17. Correlation statistics were used to obtain the correlation between metal contents measured in the sediments and *P. schlosseri* tissues selected from matched sampling sites.

The significance of these correlations was tested using Pearson correlation coefficient at 95% ($p < 0.05$) and 99% ($p < 0.01$) significance levels.

3. Results and Discussion

Tables 2-6 show the relationship between metals concentrations in the tissues of *P. schlosseri* and those in the geochemical fractions and total metal concentrations in the sediments.

Table 2 shows the correlation analysis between geochemical fractions and total metal concentrations of Cu in

Table 2. Pearson's correlation between Cu concentrations in different tissues of *P. schlosseri* and geochemical fractions in the sediments.

Parameter	Scale	Muscle	Bone	Gills	Operculum	Intestine	Liver	Cartilage
EFLE	0.235	0.263	0.234	-0.15	0.221	0.243	-0.427	0.353
Acid-reducible	0.278	0.169	0.257	-0.06	0.302	0.182	0.341	0.244
Oxidisable organic	0.619**	0.586*	0.536*	-0.214	0.541*	0.647**	0.071	0.029
Resistant	0.467	0.405	0.441	-0.193	0.477*	0.35	-0.138	0.333
Total	0.563*	0.480*	0.527*	-0.196	0.547*	0.465	-0.139	0.395

**Correlation is significant at the 0.01 level (2-tailed); *Correlation is significant at the 0.05 level (2-tailed).

Table 3. Pearson's correlation between Zn concentrations in different tissues of *P. schlosseri* and geochemical fractions in the sediments.

Parameter	Scale	Muscle	Bone	Gills	Operculum	Intestine	Liver	Cartilage
EFLE	0.217	0.028	-0.102	-0.271	-0.231	-0.457	-0.672**	-0.338
Acid-reducible	-0.1	-0.306	-0.161	-0.306	-0.295	-0.464	-0.686**	-0.403
Oxidisable organic	-0.15	-0.097	0.049	-0.21	-0.225	-0.462	-0.234	-0.423
Resistant	-0.413	-0.318	0.043	-0.152	-0.17	-0.112	0.113	-0.436
Total	-0.235	-0.277	-0.04	-0.255	-0.268	-0.428	-0.356	-0.483*

**Correlation is significant at the 0.01 level (2-tailed); *Correlation is significant at the 0.05 level (2-tailed).

Table 4. Pearson's correlation analysis, relationship between Pb concentrations in different tissues of *P. schlosseri* and geochemical fractions in the sediments.

Parameter	Scale	Muscle	Bone	Gills	Operculum	Intestine	Liver	Cartilage
EFLE	0.766**	0.349	0.742**	0.309	0.713**	0.445	0.384	0.725**
Acid-reducible	0.621**	0.112	0.606**	0.14	0.582*	0.148	0.117	0.539*
Oxidisable organic	0.608**	0.144	0.617**	0.156	0.563*	0.139	0.102	0.472*
Resistant	0.549*	0.046	0.621**	0.111	0.537*	-0.026	-0.034	0.453
Total	0.621**	0.082	0.670**	0.14	0.594**	0.056	0.032	0.535*

**Correlation is significant at the 0.01 level (2-tailed); *Correlation is significant at the 0.05 level (2-tailed).

Table 5. Pearson's correlation analysis, relationship between Cd concentrations in different tissues of *P. schlosseri* and geochemical fractions in the sediments.

Parameter	Scale	Muscle	Bone	Gills	Operculum	Intestine	Liver	Cartilage
EFLE	0.528*	-0.378	0.381	0.191	0.227	0.681**	0.529*	0.478*
Acid-reducible	-0.131	-0.166	-0.189	-0.381	-0.146	0.011	0.391	-0.162
Oxidisable organic	-0.598**	0.373	-0.582*	-0.470*	-0.243	-0.592**	-0.211	-0.407
Resistant	-0.394	0.432	-0.344	-0.333	-0.291	-0.649**	-0.098	-0.338
Total	-0.404	0.542*	-0.308	-0.3	-0.222	-0.601**	-0.235	-0.521*

**Correlation is significant at the 0.01 level (2-tailed); *Correlation is significant at the 0.05 level (2-tailed).

Table 6. Pearson's correlation analysis, relationship between Ni concentrations in different tissues of *P. schlosseri* and geochemical fractions in the sediments.

Parameter	Scale	Muscle	Bone	Gills	Operculum	Intestine	Liver	Cartilage
EFLE	-0.35	-0.505*	-0.535	-0.368	-0.45	-0.537	-0.292	-0.542
Acid-reducible	0.029	-0.283	-0.258	0.033	-0.042	-0.377	0.288	-0.508
Oxidisable organic	0.149	-0.097	-0.219	0.424	0.149	-0.368	0.694**	-0.432
Resistant	0.575*	0.884**	0.917**	0.500*	0.694**	0.938**	-0.03	0.967**
Total	0.695**	0.829**	0.775**	0.838**	0.825**	0.668**	0.480*	0.647**

**Correlation is significant at the 0.01 level (2-tailed); *Correlation is significant at the 0.05 level (2-tailed).

the sediments and Cu concentration in the tissues of *P. schlosseri*. Non-significant correlations ($p > 0.05$) were observed between Cu in all the tissues with the EFLE and acid-reducible fractions in the sediments. Significant positive correlations were observed between oxidisable-organic fraction of Cu with Cu concentrations in scale and intestine ($p < 0.01$), muscle, bone, and operculum ($p < 0.05$) with Cu oxidisable-organic fraction. This could be explained by high affinity of Cu to humic substances which are fraction of natural organic matter [18] [19] (Förstner, 1989; Pempkowiak *et al.*, 1999) and the highest percentage of bioavailable Cu which was associated with oxidisable-organic fraction of Cu in the present study. Operculum shows a positive correlation ($p < 0.05$) with the Cu resistant, the Cu concentration in scale, muscle, bone and operculum showed positive correlations ($p < 0.05$) with total Cu concentration in the sediment. The correlations between Cu in sediment and *P. schlosseri* implied that, accumulation of Cu in *P. schlosseri* was related to these fractions.

A strong affinity between Cu and organic matter, due to high complexing tendency of Cu for organic matter was reported by [20] Zhai *et al.* (2003). Therefore the bioavailability of Cu to *P. schlosseri* would depend on Cu bound to this fraction. In addition Cu in total fraction was also positively correlated with these tissues except with intestine and only operculum was correlated with resistant fraction of Cu. The correlations between Cu in sediment and *P. schlosseri* implied that, accumulation of Cu in *P. schlosseri* was related to these fractions. Non-significant correlations between EFLE and acid reducible Cu with all the examined tissues may probably be attributed to little Cu extracted from these fractions.

Pearson's correlations analysis between different geochemical fractions of Zn in the sediments and Zn in the tissues of *P. schlosseri* was presented in **Table 3**. Significant negative correlations ($p < 0.01$) were found between Zn concentration in the liver with EFLE and acid-reducible fractions of Zn in the sediment whereas Zn concentration in the cartilage was negatively correlated ($p < 0.05$) with total Zn in the sediments.

This finding is not in agreement with [21] Bochenek *et al.* (2008), who reported statistically significant positive correlation coefficients between Zn concentration in roach liver with Zn exchangeable and carbonate fractions concentrations in the sediments. The EFLE fraction is known to be the most bioavailable fraction and a better measure of bioavailable metal forms [22] (Shulkin and Presley, 2003) which is considered to have most immediate effect on benthic organisms dwelling on sediment as well as other organisms like fishes, crabs and mussels [23] (Jenne and Luoma, 1997) while the acid-reducible fraction constitutes a significant sink for heavy metals in the aquatic system.

The effect of heavy metals on aquatic organisms is controlled by the concentrations and chemical forms of the metals in water and sediment. Excessive Zn will be toxic causing mortality, growth retardation and tissue alterations [24] (Sorensen, 1991) in living organisms. Large but sub-lethal levels of Zn can induce pathological and morphological abnormalities in adult fish whereas larvae that hatched after exposure to Zn solutions displayed various malfunctions to eyes and optic capsules, and capsule jaws and branchial arch [25] (Somasundaram *et al.*, 1984).

Fish have evolved homeostatic mechanisms to maintain optimal Zn levels to meet the needs for normal metabolism and physiology without toxicity [26] (Kamunde *et al.*, 2009). Therefore, the negative correlation observed between Zn concentrations in liver with EFLE and acid-reducible fractions of Zn might suggest homeostasis of Zn in liver being a target center for metabolism. Furthermore, higher concentrations of Zn in liver were predicted to occur at sampling sites with low EFLE and acid-reducible fractions which might probably, explained the high Zn content in liver and cartilage at M. Beku and Bg. Lalang and high Zn content in intestine at S. Garam. The relationship found between chronic exposure and metal bioaccumulation in aquatic biota shows that, usually Zn content is well regulated [27] [28] (Marcovecchio and Moreno, 1993; McGeer *et al.*, 2003). The relatively large content of Zn in the tissues of the examined fish may be attributed to the percentage of Zn in the bioavailable fractions. Non-significant correlations between Zn in other tissues of *P. schlosseri* with all the geochemical fractions is further evidence that Zn is tightly regulated by fish [29] (Bury *et al.*, 2003).

Correlations analysis between different geochemical fractions of Pb in the sediments and Pb in the tissues of *P. schlosseri* was presented in **Table 4**. Significant positive correlations ($p < 0.01$ or $p < 0.05$) were found between Pb in scale, bone, operculum and cartilage with all the geochemical fractions of Pb but cartilage was not correlated with resistant fraction of Pb. Non-significant correlation ($p > 0.05$) was found between all geochemical fractions of Pb in sediments with muscle, gills, intestine and liver of *P. schlosseri*.

The correlations between Pb in sediment and *P. schlosseri* implied that Pb accumulation in *P. schlosseri* was related to metal concentrations in these fractions. It was also shown by the result of the present study that Pb concentration was generally highest in these hard tissues.

This could further be explained by the fact that, the permanent storage site of lead may be the calcified tissues, such as bones and scales, where Pb can replace calcium [30] (Coello and Khan, 1996). Non-significant correlations between all geochemical fractions of Pb in sediments with gills, intestine and liver of *P. schlosseri* might be attributed to regulatory functions of these organs. Gills and intestines are the main barriers that control absorption of metals from the environment and from contaminated preys [31] (Andres *et al.*, 2000) while liver is the major organ involved in xenobiotic metabolism in fish [32] (Romeo *et al.*, 1994). The non-significant correlation between muscles with any of the geochemical fraction of Pb might suggest muscle as poor accumulator of Pb. It has been reported [33] [34] Bradley and Morris (1986); Wagner and Boman (2003) that fish muscle as a poor accumulator of Pb. It is generally accepted that muscle is not an active tissue in accumulating heavy metals [35] (Karadede and Ünlü, 2000) due to low levels of binding proteins in the muscle tissue.

Table 5 shows correlation between different geochemical fractions of Cd in the sediments and Cd concentration in different tissues of *P. schlosseri*. Significant positive correlations ($p < 0.05$) were observed between EFLE Cd with Cd concentration in scale, liver and cartilage ($r = 0.528, 0.529$ and 0.478 respectively), but Cd in intestine shows a significant positive correlation ($p < 0.01$) with EFLE Cd fraction. High concentration of Cd in these tissues were more likely to occur at sampling sites high in EFLE Cd. Tissues concentration of Cd were generally high at sampling sites with high percentage of EFLE fraction in the present study. Oxidisable-organic fraction of Cd shows a significant negative ($p < 0.01$) correlation with scale and intestine, bone and gills were also negatively correlated ($p < 0.05$) with oxidisable-organic fraction of Cd. The resistant Cd fraction in the sediments shows significant negative correlation ($p < 0.01$) with Cd concentration in the intestine. Total Cd concentration in sediments shows significant correlation ($p < 0.05$) with muscle and significant negative correlation ($p < 0.01$) with intestine and cartilage ($p < 0.05$).

The total concentration of Cd in sediment and Cd concentration muscle were found highest at M. Beku which was in agreement with positive correlation obtained between Cd in sediment and muscle. The oxidisable-organic, resistant and total fractions of Cd were significantly and negatively correlated with Cd concentrations in different tissues of *P. schlosseri*, which might suggest that highest concentration of Cd in tissues will occur at sampling site with lowest concentrations of these fractions. Cd toxicity could be increased slightly through the presence of naturally occurring organic matter as indicated by [36] (Giesy *et al.*, 1977).

Non-significant correlations ($p > 0.05$) were found between acid-reducible fractions of Cd with all the tissues in *P. schlosseri*. It has been suggested by [37] Rautengarten *et al.* (1995) that Cd distribution is controlled by small mineral particles and organic matter, its mobility being determined by clay content. The non-significant correlation observed between acid-reducible fractions of Cd with all the tissues might be attributed to the metal concentration available to *P. schlosseri* in this fraction.

The correlation between different geochemical fractions of Ni in the sediment and Ni in the tissues of *P. schlosseri* was presented in **Table 6**. Significant negative correlation ($p < 0.05$) was observed between EFLE Ni with Ni concentration in muscle which might imply high concentrations of Ni in muscle occurred at sampling site low in EFLE Ni. The oxidisable-organic fraction of Ni showed significant positive correlations with Ni concentration in the liver. The resistant of fraction Ni showed a positive correlation ($p < 0.01$) with Ni concentration in; muscle, bone, operculum, intestine and cartilage while scale and gills showed significant positive correlation ($p < 0.05$) with resistant fraction of Ni in the sediment. Total Ni concentration in sediment shows positive correlation ($p < 0.01$) with Ni concentration in all the tissues except liver which was positively correlated at $p < 0.05$.

The positively correlations observed between oxidisable-organic, non-resistant, resistant and total Ni with Ni in different tissues of *P. schlosseri* suggest high concentrations of Ni in tissues occurred at sampling site high in these fractions. In general, Ni is present predominantly in oxidisable and residual fractions in aquatic sediments [20] [38] (Staelens, *et al.*, 2000; Zhai, *et al.*, 2003).

4. Conclusion

The results of the present study showed significant correlations between metal concentrations in some tissues of *P. schlosseri* and metals concentrations in different geochemical fractions of sediments. This indicated the ability of the fish to accumulate or regulate heavy metals in its tissues: scale, bone, operculum, intestine, liver and cartilage, therefore, the present study might suggest the use of *P. schlosseri* as a biomonitoring agent for heavy metals pollution in the west coast of Peninsular Malaysia.

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