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Relationships Between Heavy Metals in Shrimp (*Macro brachium felicinum*) and Metal Levels in The Water Column and Sediments of Taylor Creek

Opuene, K.[1]* and Agbozu, I. E.[2]

[1]Industrial Laboratory, OB/OB Gas Plant, Nigerian Agip Oil Company Ltd., PMB 35,
Omoku, Rivers State, Nigeria
[2]Department of Chemical Sciences, Niger Delta University, P.M.B. 71,
Yenagoa, Bayelsa State, Nigeria

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ABSTRACT: Heavy metals in the aquatic environment have to date originated fundamentally from naturally occurring geochemical resources. Nonetheless, this has been enhanced by anthropogenic activities resulting in pollution. Consequently, relationships and partitioning of heavy metals in the dissolved phase, suspended particulate matter (SPM), sediments and shrimp (*Macrobrachium felicinum*) were investigated in five selected sites along Taylor creek, southern Nigeria. The degree of relationships between the various metals was dissimilar in each of the investigated matrices. In the matrices studied, not several significant relationships ($P<0.05$) were obtained. Only Ni-Cd ($r=0.95$), Mn-Cd ($r=0.63$), Mn-Ni ($r=0.64$) were associated in the sediments and in SPM, Zn-Ni ($r=0.72$), which suggests that the sources were not common for both matrices. In the dissolved phase, no strong relationships ($P<0.05$) between the heavy metals were obvious. The best relationships were observed for Ni-Zn ($r=0.72$) and Cd-Pb ($r=0.65$). Partitioning coefficients (K_d) of heavy metals between dissolved phase and SPM were generally low, which is typical for fresh water ecosystems and fairly stable over the creek all through the seasons. Furthermore, the bio-concentration factors (BCFs) of *Macrobrachium felicinum* were low unlike those of other natural waters. Thus, the physical state of the aquatic ecosystem points to the fact that the heavy metals bio-accumulated by *Macrobrachium felicinum* give cause for concern when viewed in perspective to community health issues, as the communities along the creek depend directly on shrimps as a protein source.

Key words: Heavy metals, *Macro brachium felicinum*, Partitioning, sediments, SPM, Taylor creek

INTRODUCTION

Municipal and industrial discharges, urban storm-water runoff, and agricultural drainage can result in heavy metals, nutrients, pesticides, and organic wastes being transported into aquatic ecosystems. In aquatic ecosystems, contaminants are often rapidly removed from the water column via sorption processes. Given that heavy metals are not subjected to degradation processes, they tend to accumulate in benthic sediments (Baeyens *et al.*, 1998). However, heavy metals are not necessarily fixed permanently to sediments; rather they may be remobilized via chemical, physical, and biological processes (Salomons *et al.*, 1987).The pollution of aquatic ecosystems by heavy metals is a significant problem, as heavy metals constitute some of the most hazardous substances that can bio-accumulate (Rayms *et al.*, 1998; Tarifeno-Silva *et al.*, 1982; Zwieg *et al.*, 1999). Metals that are deposited in the aquatic environment may accumulate in the food chain and cause ecological damage while also posing a risk to human health (Adams *et al.*, 1992; Grimanis *et al.*, 1978).Taylor creek, a non-tidal freshwater environmental unit, is situated in Gbarain clan in the Yenagoa Local Government Area of Bayelsa State in the Niger Delta, southern Nigeria. The creek is 16km North-northeast (NNE) from the state capital of Bayelsa State

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*Corresponding author: E-mail- opuenekings@yahoo.com and enjoys the humid tropical climate characterized by the hot and wet conditions associated with the movement of the Inter-Tropical Convergence Zone (ITCZ) north and south of the equator. As a result, the study environment has two major seasons, the dry season and the wet season and experiences consistently high temperatures (about $32^{O}C$) all year round. Since temperature varies only slightly, rainfall distribution, over space and time, becomes a single most important factor (Okafor and Opuene, 2007).

In addition, the only perceptible activity in the area is oil industry activities. The Etelebou Flow Station, which is located on second order distributaries, discharges liquid effluents (either discharged untreated or with only primary treatment) into Etelebou creek, a tributary of Taylor creek. Therefore, Taylor creek is a receptor of a highly polluted creek (Okafor and Opuene, 2007). Furthermore, Taylor creek has been the source for fish, shrimps and mollusks to the residents living along the stem of the creek. It is suspected that shrimps from Taylor creek, particularly at the convergence of Etelebou and Taylor creeks will be highly contaminated with heavy metals, hence endangering the health of those who consume such shrimps. To date, information regarding the relationships and partitioning of heavy metals in the dissolved phase, suspended particulate matter (SPM), sediments and shrimps (*Macrobrachium felicinum*) from the creek and its effects is very limited. However, the relationships and partitioning of heavy metals between environmental matrices of some aquatic ecosystems have been studied by different researchers (Bryan and Langston, 1992; Hung *et al.,* 2001; Liu *et al.,* 2003; Jain and Sharma, 2001; Munksgaard and Parry, 2001; Nguyen *et al.,* 2005).In this paper, we shall provide inter-metal relationships in the dissolved phase, SPM, sediments and *Macrobrachium felicinum*, which could present information on the probable sources of the heavy metals in Taylor creek as well as their partitioning in the water column.

MATERIALS & METHODS

The study area stretched from Agbia/Nedugo to Polaku along Taylor creek, which comprised of five sampling sites. The entire stretch from Agbia/ Nedugo to Polaku is about 16km and lies between longitude $006^{O}17^{I}$ to $006^{O}21^{I}$ E and latitude $05^{O}01^{I}$ to $05^{O}05^{I}$ N respectively. Samplings of surface waters were carried out monthly for the period January to December 2006 from the sampling sites. Surface water samples were collected with nitric acid pre-rinsed 1litre plastic containers. After collection, the samples were placed in cooler boxes with ice bags whilst being transported to the laboratory and kept at about $4^{O}C$ before analysis. Surficial sediments were collected monthly from each of the five sampling sites adjacent to where the surface waters were sampled in the period January 2006 to December 2006 by the grab method using an Eckman grab sampler on 3 to 4 locations, representing approximately 2-3cm of surficial bottom sediments, within each sampling site, and wrapped with aluminium foil to avoid contamination, frozen and taken to the laboratory. Individual site (sediment) sample was thawed and air-dried at ambient temperature and pulverized to $< 50\mu m$ using a shatter-box grinding mill. Samples of shrimp (*Macrobrachium felicinum*) were collected each month from January to September 2006 from the sampling sites as well. All samples of *Macrobrachium felicinum* were washed three times: in the freshwater, in distilled water and in freshwater again to remove epifauna.

The water samples, after filtration with 45 μm Whatman filter paper, were acidified and pre-concentrated. The filtrates were digested with a mixture of 10mL of conc. nitric acid and 2mL of conc. perchloric acid respectively. Two grams of air-dried sediment samples were also weighed using a high precision microscale for each case, moistened with water and put into a 50mL conical flask. To each weighed sample, 10mL of conc. nitric acid and 2mL of conc. perchloric acid were added and heated with a water bath to near dryness. The mild acid treatment was optimal to digest organic matter with associated metals so as to extract metals linked by adsorption to the sediments, which together represent bio-available or leachable metals of the surficial sediments. Each solution was allowed to cool and subsequently filtered into a 50mL volumetric flask. The treatment of *Macrobrachium felicinum* samples was based on established procedure (Agbozu and Opuene, 2008). Before analysis, the *Macrobrachium felicinum* samples were dehydrated to a constant weight using an oven at $65^{O}C$ for 20hours and individual whole sample pulverized to a uniform particle size. Two grams of pulverized weight were weighed using a high precision microscale and put in a digestion flask and digested with a mixture of 10mL of conc. nitric acid and 2mL of conc. perchloric acid. The contents of the flask was, for each case, digested gently and slowly, by heating in a water bath until the contents got to near dryness. It was then set aside to cool. The digest was filtered into a 50mL volumetric flask, and made up to mark with distilled water.

Following acid digestion, all samples were analysed for five elements: Cd, Pb, Ni, Mn and Zn by flame atomic absorption Spectrophotometry using Buck Scientific Model 200A Spectrophotometer, equipped with a high sensitivity nebulizer. Calibration of Buck Scientific Model 200A Spectrophotometer was performed before every run by successive dilution of a 100mg/L Multi-Element Instrument Calibration Standard solution (Fisher Scientific) that was in a range covering the concentration levels in the analysed samples. For each batch of elemental analyses, intra-run quality insurance standard (1mg/L, Multi-Element Standard Solution, Fisher Scientific) was checked for reading variation and precision of every 10 samples. Internal blanks were used to assess any background contamination originating from sample manipulation and preparation. Blanks were processed exactly as respective regular samples as well (Okafor and Opuene, 2007). The accuracy of sample manipulation for the heavy metals was checked using samples of CASS-4 (seawater), PACS-2 (sediment) and DOLT-3 (organism tissue) Matrix Certified Reference Materials with known concentration for certain metals and were found to be ≥89% for Cd, Pb, Ni, and Zn (Cantillo and Calder, 1990).The relationship between inter-metal correlations in the segments of the creek was tested using Pearson Product Moment Correlation Coefficient. Relationships between the dissolved phase, particulate phase or suspended particulate matter (SPM) and *Macrobrachium felicinum* were discussed via the corresponding partition coefficients and bio-concentration factors (BCF). Bio-magnification, the enrichment (positive value) or depletion (negative value) of a higher trophic level versus food was also calculated. All statistical analyses were done using Analysis Toolpak soft- ware, with significance based on an α of 0.05 (Zar, 1996).

RESULTS & DISCUSSION

The assessment of heavy metals and their correlations in Taylor creek reflected the degree of pollution, which is considered by many regulatory agencies to be one of the principal risks to the aquatic ecosystem (Table 1). The mean levels of Cd, Pb, Mn, Ni and Zn in the dissolved phase were higher than the interim standards for aquatic life and domestic use (FEPA, 1991; WHO, 1989). Based on the guidelines, direct use of water from the creek without treatment may aggravate poor health of sensitive groups. For example, the criterion for Pb in water for domestic use is 0 to 1.70μg/L (FEPA, 1991). At levels > 100μg/L, possible neurological damage in foetuses and young children may occur (Fatoki *et al.*, 2002). For some of the metals studied in the surface sediments, except for Cd, the levels were higher than the metal levels in the sediments of Montevideo Harbour, Uruguay (Muniz *et al.*, 2003). Although, it is apparent that the concentration levels of the metals may be due, partly, to catchments in-washings (Ibok *et al.*, 1989), oil industry activities are also implicated (Nwadinigwe and Nwaorgu, 1999). Besides, the study sites are located in the lower reaches of Taylor creek suggesting that inputs from Etelebou creek, a tributary of Taylor creek, may be an influencing factor. In spite of the levels of heavy metals in the sediments, we can deduce that the sediments present concentrations that were at the Persaud et al. (1992). Severe Effect Level and may cause adverse biological effects except for Ni and Zn respectively. Absolute values of K_d in Taylor creek, as presented in (Table 2), are defined as the ratio of the particulate metal concentration (mg/kg) over the dissolved metal concentration (mg/L):

$$K_d = \frac{[\text{particulate metal concentration}]\ (\mu g/kg)}{[\text{Dissolved metal concentration}]\ (\mu g/L)}$$

The calculated results show that the log (K_d) values for Cd, Pb, Ni, Mn and Zn were comparatively constant and low unlike the log (K_d) values of the Scheldt Estuary and Lake Balaton (Baeyens *et al.*, 1998; Nguyen *et al.*, 2005). And also, K_d values in Lake Balaton ranged from 4.3 to 6.0 in June 2000 and from 3.3 to 6.4 in September 2001 due to the influence of stormy weather conditions (Nguyen *et al.*, 2005), which shows variations between the seasons. Conversely, the seasonal and spatial distributions of K_d in Taylor creek were rather stable for the metals studied. The stable K_d values of the metals may be due to the nature of the dissolved metals in the creek.

Table 1. Mean levels of heavy metals in SPM, dissolved phase, sediments and Macrobrachium felicinum

	Parameter (Mean ± SD)				
Matrix	**Cd**	**Pb**	**Ni**	**Mn**	**Zn**
SPM, µg/g dry weight	0.0059 ± 0.0076	0.019 ± 0.02	0.067 ± 0.023	0.09 ± 0.303	0.083 ± 0.181
Dissolved phase, µg/mL	0.0023 ±0.007	0.0079 ± 0.03	0.026 ± 0.03	0.035 ± 0.354	0.032 ± 0.223
Sediments, µg/g dry weight	2.870± 2.599	115.16± 251.3	10.18± 8.732	266.92± 287.22	107.73± 98.61
Macrobrachium felicinum, µg/g dry weight	0.08±0.10	0.350±0.08	0.156±0.06	3.736±1.059	2.516±0.46

Table 2. Absolute values of log (*K*d), log (BCF) and log (BMF)

Metal	log (*K*d)	log (BCF)	log (BMF)
Cd	0.41	1.40	-0.35
Pb	0.33	1.75	0.48
Mn	0.41	2.45	1.51
Zn	0.41	2.73	1.34
Ni	0.41	1.68	0.12

For the levels of Cd, Pb, Ni, Mn and Zn, inter-metal relationships appear to be different in the environmental matrices (Tables 3-7). While, inter-metal relationships exist between Cd-Ni (*r*=0.95) and considerable correlation between Cd-Mn (*r*=0.63) and Ni-Mn (*r*=0.64) for the sediments, which were comparable to correlation coefficients reported elsewhere (Hung *et al.*, 2001; Liu *et al.*, 2003), there were no numerous correlations between the heavy metals in SPM. However, the correlation matrix for SPM shows significant inter-metal relationships (*P*<0.05) between Ni-Zn (*r*=0.72). Furthermore, considerable correlations between Cd-Pb (*r*=0.50) was also observed for SPM. According to Nguyen et al. (2005), a considerable number of significant correlations in SPM of Lake Balaton were observed between metals. However, Jain and Sharma (2001) found very strong correlations on similar metals from the Hindon River in India. In addition, it was observed that all particulate metals are negatively correlated with the amount of SPM that was similar to the trend in Hindon River (Jain and Sharma, 2001), which may be due to a dilution process of permanently suspended matter, loaded in heavy metals coupled with the temporary re-suspension of bottom sediments less contaminated by metals (Nguyen *et al.*, 2005). Strong correlations between Ni and Zn in the SPM and dissolved phases suggest similar partitioning mechanism in the creek. The significant correlations between Ni-Zn in the two phases also confirm the impact of anthropogenic input on the environmental matrices. In the dissolved phase, no strong correlations between the heavy metals were obvious. The best correlations, significant at *P*<0.05, were observed for Ni-Zn (*r*=0.72) and Cd-Pb (*r*=0.65). This implies that the cycling of the correlated metals may be associated. Also significant but relatively weak correlations (*r*≥0.50) were observed between Pb-Ni. Furthermore, only few correlations between dissolved heavy metals have been reported (Munksgaard and Parry, 2001).

In view of total metal concentrations in the water column as the sum of dissolved and particulate metal concentrations, correlationsbetween these metals are much better than in individual phases (Table 7). Again Ni-Zn correlation is recognized as one of the strongest with a correlation coefficient *r*= 0.72. Despite

Table 3. Pearson product moment correlation coefficients between particulate metal levels

	Cd	Pb	Ni	Mn	Zn
Cd	1.00				
Pb	0.47496	1.00			
Ni	0.275733	-0.46188	1.00		
Mn	0.247107	0.204431	0.12518	1.00	
Zn	-0.13885	-0.86775	0.720149	-0.11465	1.00

Table 4. Pearson product moment correlation coefficients between dissolved metal levels

	Cd	Pb	Ni	Mn	Zn
Cd	1.00				
Pb	0.65117	1.00			
Ni	0.274452	0.499155	1.00		
Mn	0.246703	0.281182	0.12561	1.00	
Zn	-0.14088	0.249695	0.719814	-0.11465	1.00

Table 5. Pearson product moment correlation coefficients between metal levels in sediments

	Cd	Pb	Ni	Mn	Zn
Cd	1.00				
Pb	-0.04827	1.00			
Ni	0.949162	-0.18831	1.00		
Mn	0.63202	0.40707	0.642074	1.00	
Zn	0.246692	0.120621	0.305322	0.393387	1.00

Table 6. Pearson product moment correlation coefficients between metal levels in Macrobrachium felicinum

	Cd	Pb	Ni	Mn	Zn
Cd	1.00				
Pb	0.849859	1.00			
Ni	0.894339	0.745516	1.00		
Mn	-0.45712	-0.64924	-0.49846	1.00	
Zn	0.585341	0.809448	0.263584	-0.36632	1.00

Table 7. Pearson product moment correlation coefficients between total metal levels in the water column

	Cd	Pb	Ni	Mn	Zn
Cd	1.00				
Pb	-0.79782	1.00			
Ni	0.38147	-0.42762	1.00		
Mn	0.337201	-0.09499	0.769786	1.00	
Zn	0.062601	-0.34928	0.720839	0.640942	1.00

insignificant correlations of Mn with other metals in the dissolved or SPM phase, total Mn correlates between these metals are much better than in individual phases (Table 7). Again Ni-Zncorrelation well with Ni (*r*= 0.77). One other significant correlation is between Mn-Zn (*r*= 0.64). The fact that total Pb was correlated with SPM levels (*r*=0.97) implies a strong dependence of Pb on SPM content in the water column, which was also observed in Bynoe river (Munksgaard and Parry, 2001). The strong dependence of total Pb on the amount of SPM reflects the importance of the particulate phase. This therefore indicates that the more the SPM in the water column, the higher the volumetric particulate metal level. In Taylor creek, the low mean depth and the preponderance of silt in the sediments indicates that the bottom and surficial sediments are easily disturbed in the flood time of the year and resuspended, thus increasing the quantity of SPM. Owing to such situation, Pb, Mn and Zn metals were observed to be remobilized that impacted the dissolved phase, which was confirmed by the correlation coefficients for the metals: Pb in sediments was significantly correlated to Pb in the dissolved phase (*r*=0.998), Mn in sediments was extensively correlated to Mn in the dissolved phase (*r*=0.90) and Zn in sediments was considerably correlated to Zn in the dissolved phase (*r*=0.71) respectively.

BCF and BMF are metal- and organism-dependent. BCF represents the uptake of metals into an organism from the surrounding water alone. The accumulation process involves the biological sequestering of metals that enter the organism through respiration and epidermal (skin) contact with the metals. The sequestering results in the organism having a higher concentration of the metals than the concentration in the organism's surrounding environment. Furthermore, BMF refers to bioaccumulation of metals up the food chain by transfer of the metals in smaller organisms that are food for larger organisms in the chain. It generally refers to the sequence of processes that result in higher concentrations in organisms at higher trophic levels. These processes result in an organism having higher concentrations of the metals than is present in the organism's food. BCF and BMF were calculated as follows:

$$\text{BCF}=\frac{[\text{Concentration of metal in } \textit{Macrobrachium felicinum}]\ (\mu\text{g/g})}{[\text{Concentration of metal in dissolved phase}]\ (\mu\text{g/mL})}$$

$$\text{BMF}=\frac{[\text{concentration of metal in } \textit{Macrobrachium felicinum}]\ (\mu\text{g/g})}{[\text{Concentration of metal in SPM}]\ (\mu\text{g/g})}$$

Log (BCF) values of Mn and Zn in *Macrobrachium felicinum* were the highest, whereas the lowest one was found for Cd. The ecological distribution of the log (BCF) values was, for all the metals, moderately stable over the creek (Table 2). All log-transformed BMF values in the creek were positive except for Cd, which indicates that the metal concentrations were larger in *Macrobrachium felicinum* than in SPM. Conversely, Nguyen et al. (2005) reported that all log-transformed BMF values in Lake Balaton are negative, which means that the metal concentration was larger in suspended matter than in the organism studied. For Ni, positive but low values (log (BMF) =0.12) reveal the degree of bio-magnification of this metal across the creek. The absolute log (BMF) values of heavy metals in Taylor creek can therefore be ranked in order of decreasing magnitude: Mn (1.51) > Zn (1.34) > Pb (0.48) > Ni (0.12) > Cd (-0.35). This sequence indicates that toxic metals such as Pb and Zn are undergoing significant bio-reduction from the SPM to *Macrobrachium felicinum* (Nguyen *et al.*, 2005).

CONCLUSIONS

Inter-metal relationships were dissimilar in the dissolved phase, SPM and sediments of Taylor creek. The amount of strong and significant relationships was better in the sediments than in dissolved phase and SPM respectively. Strong relationships between Ni and Zn in the dissolved phase and SPM suggest analogous partitioning mechanisms in Taylor creek. Significant relationships between the metals in the phases point to the fact that the sources of the heavy metals are not common. Correlations of total metal levels were also better than the individual phases. Besides, total Pb was correlated with SPM levels, which shows a strong dependence of Pb on SPM content in the water column. Furthermore, Pb, Mn and Zn metals were observed to be remobilized that impacted the dissolved phase. Therefore, the physical state of the aquatic ecosystem points to the fact that the heavy metals bio-accumulated by *Macrobrachium felicinum* gives cause for concern when viewed in perspective to community health issues, as the communities along the creek depend directly on shrimps as a protein resource.

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