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Eco-partitioning and indices of heavy metal accumulation in sediment and *Tilapia zillii* fish in water catchment of River Niger at Ajaokuta, North Central Nigeria

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In this study the distribution and accumulation indices of some heavy metals in sediments and *Tilapia zillii* fish in freshwater catchment of River Niger by Ajaokuta Steel Company (ASC), North Central Nigeria were investigated. Water, bottom sediments and *Tilapia zillii* fish samples were collected upstream and downstream of the drainage column by ASC, Ajaokuta. The sample were digested according to standard methods and analysed for Cd, Mn, Cr, Ni, Cu, Zn and Pb using flame atomic absorption spectrometer. Accumulation indices or factor (AI or AF) of the investigated heavy metals were defined using the ratio of mean concentration C_o in component/organism and that in the surrounding water C_w at steady state ($AI/AF = C_o/C_w$). Sediment accumulation indices (AI) of the metals were: Cd, 5.4; Mn, 3.4; Cr, 1.6; Ni, 12.5; Cu, 1.6; Zn, 25.9 and Pb, 411.6, while the AI of the metals in fillets of *T. zillii* were Cd, 3.0; Mn, 2.1; Cr, 1.6; Ni, 5.1; Cu, 4.6; Zn, 3.2 and Pb, 14.0. Seasonal climate changes induces little marginal or no changes in the AIs of the metals except for Pb (841.5, 411.6) and Cd (11.6, 5.4) in sediment, and Pb (32, 14) and Cd (5.3, 3.0) ($p<0.05$) in fish fillets. Thus significant changes in metal AIs may be the consequence of their concentration levels in the aquatic ecosystem. Therefore, accumulation indices or factors may be an estimate of ecosystem status, and may be a useful tool for monitoring and predictive risk assessment (MPRA) purposes.

Key words: Partitioning, accumulation indices, heavy metals, fish fillet, bottom sediment.

INTRODUCTION

The sustenance of aquatic ecosystem is a function of the quality and health of the water catchment, the physico-chemical composition and the balance in the dynamics of the ecosystem structure (Waldichuk, 1977). The quality of water in streams, rivers and lakes, as well those of coastal and marine water is hampered by the presence of contaminant substances such as heavy metals, when present in concentrations beyond natural levels. The notoriety of heavy metals arises from its non degradable character which results in its persistence. The tendency for the accumulation of heavy metals beyond tolerable concentration in biotic and non-biotic functions, gives rise to its bio-toxic tendencies (Sieckhaus, 2009).

The efficiency of the dynamics of material circulation relies on the type, nature and sources of input from the different release points. Orlob (1975) reported that

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circulation was an important determinant of ecosystem response to material input into aquatic environment. Partitioning and accumulation of heavy metals in the different compartments of an aquatic environment is therefore a function of the ecosystem response to contaminant loads (Zhou et al., 2008). The response mechanism of aquatic ecosystem to input of contaminants is influenced such factors as, temperature zoning, vertical stratification, quality of inflow from the arterial networks/tributaries, ecosystem flora and fauna population, primary productivity, turbidity, light penetration, environment/habitat loss or creation, and hydraulic residence time variation resulting from sedimentation and the underlying water geology (Okabe et al., 1993; Leclerc et al., 1995; Shen et al., 1995; Bailey and Hamilton, 1997; Hamilton and Schladow, 1997; Schladow and Hamilton, 1997; Cook and Burkhard, 1998).

The partitioning of heavy metal in water follows series of complicated pathways including microbial, flora and fauna uptake, siltation/sedimentation etc., and this may lead to accumulation in aquatic biota and in different compartments of the water catchment (Moustafa, 2000). Heavy metals accumulation depends on metals availability and concentration, the rate of metabolism of bioaccumulative chemicals in biotic function, and or their stabilization in non-biotic function such as sediments (Mitsch et al., 1989). The accumulation of heavy metal pollutants in fish and other aquatic foods was reported to be a potential route of human exposure (ATSDR, 1999). Thus, high concentration of heavy metals in fresh water rivers present a major safety and health risk to humans and higher heterotroughs, who depend on such system for food resources.

The accumulation of chemical substances is an intrinsic property, and this determines their potential environmental hazard (Schlechtriem et al., 2012). Assessing the accumulation and or bioaccumulation potential of heavy metals is crucial to environmental and human risk assessment, because it is one of the main features required for environmental monitoring. According to Adolfsson et al. (2012), modern chemical legislation requires measurement of bioaccumulation factor (index) of large number of chemicals in fish and other edible aquatic organism. Data on accumulation indices needed for quantitative measurement of heavy metal partitioning between components of freshwater aquatic ecosystems, in order to define their exposure risk, as well as ecosystem health and status, especially that of River Niger are very limited. The indices of accumulation of contaminants such as heavy metals, may be leading in deriving pollution predictive indices (PPI) which can defines contaminant status of and aquatic ecosystem. Guidelines for the determination of bioaccumulation factor (index of bioaccumulation) are listed in the Organization for Economic Cooperation and Development (OECD) technical guideline (TG) 305.

In this study the distribution and accumulation indices (factors) of some heavy metals in sediments and *Tilapia zillii* fish in fresh water catchment of River Niger ecosystem by Ajaokuta Steel Company, North Central Nigeria were investigated. This is in order to define the dynamics of heavy metals partitioning and accumulation in the column segment, and the likely ecosystem response to input of heavy metals from the activities of Ajaokuta Steel Company.

MATERIALS AND METHODS

Description of study area

Ajaokuta is situated along the lower River Niger drainage at about N 07.508848°, E 006.692085° (Figure 1). Lower River Niger stretches from below the Niger-Benue confluence at Lokoja North Central Nigeria flowing between the geo-reference coordinate N 07.747450°, E 006.756990^v northern latitude, down toward the equatorial zone N 04.394008°, E 007.085884°, down to the Niger Delta where it delivers its content via Bonny estuary into the Atlantic Ocean. The inter-annual mean discharge of 5589 m^3 s^{-1} was recorded between, 1946 to 1992. Historically, the minimum and maximum drainage flows ever measured in River Niger were 500 and 27,600 m^3 s^{-1} respectively (Hubert, 2000; Abrate et al., 2013). A number of tributary rivers contribute to drainage volume of the Lower River Niger, with the largest average contribution of 3500 m^3 s^{-1} from River Benue. The mean water residence time in the reservoir is around 40 days while the water temperature in the reservoir generally ranges between 27 and 34°C (Hubert, 2000). A number of riverside communities are found along the lower River Niger drain, whose occupations include fishing, animal farming and other agriculture activities. A power generation plant and a steel production plant are sited along the bank of the Lower River Niger at Geregu and Ajaokuta respectively.

Sample collection

Samples were collected up-stream and down-stream, along River Niger column by Ajaokuta Steel Company Complex located on N 07.508448°, E 006.692500° (altitude range 74 to 187 m) at Ajaokuta, North Central Nigeria. Three sample types consisting of one hundred and sixty samples each of water samples, bottom river sediment and *T. zillii* (Redbelly Tilapia) fish species were collected over 24 months period spanning between period 2004 and 2005.

Sample preparation

Sediment samples were sorted to eliminate pebbles and coarsy materials, and air-dried under ambient conditions inside the laboratory for seven-two hours. Fish samples that is, *T. zillii* fish samples were rinsed with distilled water and dried in hot air oven at temperature of 80°C to constant weight (Zheng et al., 2007). The dried soil and sediment samples were pulverized sieved through a nylon sieve of 2 mm mesh size. The dried fish were air cooled to room temperature, and sorted to remove bones from the fish muscles (fillets). The fish fillets were blended using National blender with stainless steel cutters to fine particle sizes suitable for digestion.

Samples digestion

Water samples

The method prescribed by the American Public Health Association

Figure 1. Map showing the Lower River Niger Drainage flow from below Lokoja through Ajaokuta down to the massive Niger Delta (Source: Google Maps).

(APHA, 1998) was used. The water samples were mixed from which about 100 ml each of the water samples was measured into separate clean 250 ml beakers. The samples were digested by heating on hot plate to volume of about 20 ml. The solutions of digests were filtered into 100 ml volumetric flasks, and made up mark with distilled water.

Sediment samples

Five gram each of sediment samples was weighed into 250 ml nitric acid pretreated teflons beakers. 50 ml and 2 M nitric acid analar grade reagent was added to each beaker and heated in a water bath for two hours (Oniawa, 2000). The resulting sample digests was filtered into 100 ml volumetric flasks and made up to 100 cm^3 mark with distilled water.

Fish samples

One gram dry weight of each fish samples muscles (fillets) were weighed into 100 ml kjeldahl flasks into which 1 ml concentrated nitric acid analar grade reagent was added. These were heated in water bath at 80°C for 3 h to ensure complete digestion (UNEP, 1984). The resulting sample solutions were filtered into 100 ml volumetric flasks and made up to 100 cm^3 mark with distilled water.

RESULTS

Concentrations of heavy metals in water, sediment and fish fillet of *T. zillii*

The concentration levels (mg/L) of heavy metals in water samples collected from River Niger were ranged; Mn, 1.74 to 8.37 (3.85 ± 0.93); Zn, 0.98 to 4.82 (2.72 ± 0.57); Cu, 0.58 to 4.50 (2.17 ± 0.73); Cr, 0.53 to 4.09 (2.08 ± 1.27); Ni, 0.48-1.12 (0.78 ± 0.12); Cd, 0.02-0.13 (0.05 ± 0.02); and Pb, 0.01 to 0.16 (0.03 ± 0.02) (Table 1). The results showed fairly stable metal concentration in water, with distribution sequence in the order Mn > Zn > Cu > Cr > Ni > Cd > Pb. The concentration of metals were within the fresh water guideline requirement for aquatic life recommended by the Canadian Council of Ministers of Environment (CCME, 1999).

The concentrations (mg/kg) of heavy metals in sediments from River Niger were: Zn, 36.64 – 96.23 (70.70 ± 10.68); Mn, 4.97 to 21.77 (13.24 ± 2.04); Pb, 8.84 to 17.52 (12.35 ± 1.14); Ni, 2.65 to 18.61 (9.67 ± 2.91); Cu, 0.89 to 8.21 (3.58 ± 1.32); Cr, 0.48 to 13.08

Table 1. Mean concentration levels of heavy metals detected in water, sediments and fish samples.

Sample type	Cd	Mn	Cr	Ni	Cu	Zn	Pb
Water							
Range	0.02 – 0.13	1.74 – 8.37	0.53 – 4.09	0.48 – 1.12	0.58 – 4.50	0.98 – 4.82	0.01 – 0.16
Mean concentration ± standard deviation (mg/L)	0.05±0.02	3.85±0.93	2.08±1.27	0.78±0.12	2.17±0.73	2.72±0.57	0.03±0.02
Fresh water guideline (CCME, 1999)	0.06	4.0	0.01 – 5.00	1.4	0.05 – 2.00	5.0 – 15.0	0.05 – 0.10
Sediment							
Range	0.07 – 0.62	4.97 – 21.77	0.48 – 13.08	2.65 – 18.61	0.89 – 8.21	36.64 – 96.23	8.84 – 17.52
Mean concentration ± standard deviation (mg/kg)	0.27±0.07	13.24±2.04	3.38±0.76	9.76±2.91	3.58±1.32	70.70±10.68	12.35±1.14
GESAMP guideline (1984)	0.11	770.00	-	-	33.00	95.00	19.00
Fillets of *T. zillii* fish							
Range	0.06 – 0.25	1.32 – 17.59	1.78 – 5.37	1.36 – 6.20	4.78 – 19.34	4.09 – 8.93	0.21 – 0.68
Mean concentration ± standard deviation (mg/kg)	0.15±0.03	8.29±3.60	3.38±0.65	4.03±0.94	10.10±2.95	8.95±1.43	0.42±0.10
FAO/WHO Guideline (1983)	2.00	1250	50	-	30.00	100.00	2.00

(3.38 ± 0.76) and Cd, 0.07 to 0.62 (0.27 ± 0.07). The distribution of the investigated metals in sediment were in the order Zn > Mn > Pb > Cu > Cr > Ni > Cd. These concentrations are within typical levels for uncontaminated sediments as suggested by the Joint Group of Experts on the Scientific Aspect of Marine Pollution (GESAMP, 1984).

T. zillii fish fillets heavy metals concentrations (mg/kg) in fish harvested from River Niger at Ajaokuta, were as follows: Cu, 4.78 to 19.34 10.10 ± 2.; Zn, 4.09 to 8.93 8.95 ± 1.43; Mn, 1.32 to 17.59 8.29 ± 3.60; Ni, 1.36 to 6.20 4.03 ± 0.94; Cr, 1.78 to 5.37 3.38 ± 0.65; Pb, 0.21 to 0.68 0.42 ± 0.10; Cd, 0.06 to 0.25 0.15 ± 0.03. Therefore, *T. zillii* contains safe levels of heavy metals, and this is indicative of the heavy metal levels of their habitat in River Niger at Ajaokuta. The concentration distribution sequence of the investigated metals in sediment were in the order Cu > Zn > Mn > Ni > Cr > Pb > Cd.

DISCUSSION

Concentration of heavy metals in river Niger

Substantial amount of contaminants including heavy metals from anthropogenic sources are deposited into surface water via the atmosphere and seasonal runoffs from over land (George et al., 2004). Water in streams and rivers having contaminants levels surpassing the natural or allowable threshold concentrations were reported to impact poor or fair biological condition on the streams (Boward et al., 1999). Aquatic and terrestrial organisms may be exposed to these substances through aquatic food consumption or from other use of such contaminated streams/rivers.

Aquatic ecosystems with high ecological sensitivity may respond in such a way that there may be sharp changes in ecological functions and or disruption which may lead to habitat loss, although this depends on the magnitude of impact on the physico-chemical and biological conditions. Some heavy metals are taken up and bio-accumulate in higher concentrations, thus producing higher concentration of the substances in the organism than in its environment. This may lead to significant change in the ecosystem structure, where surviving species may have their biomass contaminated, which may result in composition change. Most of these changes may be the result of the accumulation of heavy metals beyond tolerable or health limits.

Early signs of aquatic ecosystem degradation resulting from the accumulation of some heavy metals are usually obvious in biological functions. This may include fish kill, fish/fauna migration, antagonistic or synergistic effects on primary productivity and the depletion of other biological functions. Scott et al. (1986) reported that environmental perturbations including habitat alteration, and the degradation of the intra-gravel

Table 2. Accumulation factor of heavy metals concentrations in sediment and fish fillets of *Tilapia zillii* with respect to heavy metals concentrations in water of River Niger at Ajaokuta

Environment	Accumulation medium	Metal	Accumulation factor	
			Wet season	**Dry season**
	Sediment	Cd	5.4	11.6
	Sediment	Mn	3.4	2.7
	Sediment	Cr	1.6	5.4
River Niger	Sediment	Ni	12.5	9.3
	Sediment	Cu	1.6	1.6
	Sediment	Zn	25.9	24.8
	Sediment	Pb	411.6	841.5
	T. zillii Fish	Cd	3.0	5.3
	T. zillii Fish	Mn	2.1	1.4
	T. zillii Fish	Cr	1.6	3.4
River Niger	*T. zillii* Fish	Ni	5.1	3.8
	T. zillii Fish	Cu	4.6	3.3
	T. zillii Fish	Zn	3.2	4.0
	T. zillii Fish	Pb	14.0	32

environment appeared to have a greater impact on fish species than on the non biotic function of the aquatic ecosystem.

Accumulation of heavy metals *T. zillii* fish fillets and bottom sediment

The indices of accumulation (AI) or accumulation factor (AF) of contaminants may be used to define the health, safety and potential risk of exposure of aquatic ecosystems resources and components. Derivations of AIs for different contaminants facilitate the possibility of predicting the status of an environment based on comparison of monitoring indices with baseline index. Water quality initiative (WQI) of U.S. EPA illustrates the importance of the linkage between sediments and the water column and its influence on exposure of all aquatic biota (Cook and Burkhard, 1998). Heavy metals do not remain for long in water, hence they re-distributed by partitioning between water, biotic and non-biotic functions of the aquatic ecosystem, especially sediments, fish, microphytes etc. The distribution of metals between sediment and water column and between biological species such as *T. zillii* and water column can be characterized with sediment – water or fish – water quotient index (accumulation factor). Accumulation index or factor (AI or AF) is defined as the ratio of contaminants mean concentration C_o in component/organism and that in the surrounding water C_w at steady state, via all routes of exposure ($AF = C_o/C_w$) (Walker et al., 2003).

The concentrations of heavy metals in sediments of River Niger at Ajaokuta were found to be higher than in its water. Sediment heavy metals index ratio (AI) or accumulation factor (AF) with respect to heavy metals concentrations in water samples (Table 2) were: Cd, 5.4; Mn, 3.4; Cr, 1.6; Ni, 12.5; Cu, 1.6; Zn, 25.9 and Pb, 411.6, and Cd, 11.6; Mn, 2.7; Cr, 5.3; Ni, 9.3; Cu, 1.6; Zn, 24.8 and Pb, 841.5 during wet and dry season respectively. This showed that sediment concentrate Pb several folds, in the order of 411.7 to 841.5 more than Pb levels in water. Zinc, Ni, Cd, Cr and Mn also accumulated 24.8 to 25.9; 9.3 to 12.5; 5.4 to 11.6 1.6 to 5.3; and 2.7 to 3.4 folds respectively in sediments, compared to their levels in water from the same river, while Cu in sediment was accumulated at approximately two fold of its concentrations in water. This implies that, the concentration levels of heavy metals found in sediments were higher than in water (sediment > water) in the same hydrological environment as expected, that is, in River Niger.

T. zillii fish also accumulated several folds concentrations of heavy metals in their muscles (fillets) compared with metals concentration levels in water. The accumulation factor of heavy metals of *T. zillii* fish in Rivers Niger were: Cd, 3.0 to 5.3; Mn, 1.4 to 2.1; Cr, 1.6 to 3.4; Ni, 3.8 to 5.1; Cu, 3.3 to 4.6; Zn, 3.2 to 4.0; Pb, 14.0 to 32.0. There was also several fold accumulation of Pb in *Tilapia zillii* fish, 14.0 to 32.0 folds higher compared with Pb levels in water of River Niger at Ajaokuta. Nickel, Cu, Zn, Cd, and Mn accumulated 5.1; 4.6; 3.2; 3.0 and 2.1 folds respectively than in water in River Niger, with Cu at 1.6, approximately one and a half fold its levels.

Indices of heavy metal accumulation may be generic and species specific as well as environment specific. This takes cognizance of the organism uptake/intake, metabolism and chemical composition. Cook and Burkhard (1998) reported that the degree to which quantitative difference in accumulation of heavy metals may be attributed to a particular ecosystem conditions, apart from the influence of organic carbon which determines the bioavailability variables. Also the degrees of depuration of heavy metals in different fish and organisms, affects their retention capacity. Variation in metabolism and mechanism of depuration of different fish accounts for the species differences in retention or holding of heavy metals.

In general, the AIs of heavy metals in sediment were higher than AIs of heavy metals in fish, while metal levels in the water column were the least in water of River Niger.

Seasonal influence on heavy metal accumulation in aquatic ecosystem

The effect of seasonal climate change was evaluated by comparing the observed indices of heavy metal accumulation in the bottom sediment and fillets of *T. zillii* fish samples during dry and wet season. The accumulation indices of the concentration levels of the heavy metal in fillets of *T. zillii* and bottom sediment of River Niger were slightly higher during dry season water than in wet seasons except for Pb and Cd. This could be the result of stability in flow conditions and less water turbulence associated dry season low water volume in the river drainage.

Deus et al. (2013) reported that water quality is seasonally variable especially with properties including water temperature, dissolved oxygen, and contaminants load. This is because of the variations and distributions of the frequency and the percentage of extreme precipitation to the daily precipitation data of annual rainfall (Zhang et al., 2008). Wang et al. (2008) however, noted that annual extreme precipitation changes and stream flow processes resulted in little changes in terms of various indices. Aside from the sediment AI for Pb (841.5) and Cd (11.6) during dry season, which was twice ($p<0.05$) the AI (411.6) and (5.4) respectively calculated for wet season, the calculated AIs for the investigated metal were not significantly different ($p>0.05$) during dry and wet season. Similarly, the AI for Pb (32) and Cd (5.3) in fish fillets during dry season was more than twice the Pb AI (14) and Cd (3.0) respectively calculated for wet season. Mitsch et al. (1989) reported that marginal changes can affects seasonal patterns of nutrient uptake and release especially during the growth season of some aquatic biota, where uptake and immobilization by microflora, microfauna and macrophytes retain contaminants. However the dieback of aquatic plants releases contaminant back to the water column through decomposition. However, the relationship between contaminants discharged in water and aquatic ecosystem is however complex.

Uncertainty in accumulation indices

Although study results showed that there was no significant change in AI of the investigated metals except for Pb and Cd, the use of the accumulation indices for monitoring and predictive risk assessment (MPRA) purposes may be limited by some degree of uncertainty based on the degree of sensitivity of the aquatic ecosystem. For instance, the uncertainty associated with the use of fish AI may arise is as a result of their migratory nature, and their environment and site specific variation in metabolism and depuration. This may leaves the AI of fish with some uncertainty especially in its use for MPRA. Similarly changes in natural composition of sediment (e.g. reduction in sediment clay mineral and organic carbon) resulting in weak metal stabilization. This may lead to changes in AIs.

Conclusion

The result showed that the accumulation indices of the investigated heavy metals in River Niger by Ajaokuta Steel Company, Ajaokuta were nearly consistent through the study period, except those for Pb and Cd which showed significant seasonal variation. Although the status an aquatic ecosystem can be determined by compliance monitoring, the result suggest the use of accumulation indices and or factors as a comprehensive status and risk based tool, required for MPRA purposes, especially when ASC comes into operation. This is because of the comprehensive inclusion of contaminant exposure route vis-a-vis metal partitioning into different compartments and biological assimilation. Accumulation of heavy metals in aquatic ecosystem depends on the concentration levels, and the partitioning of the metals in water.

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