Pollution Status of Heavy Metals in Sediments, Water and Biota of Kalabari Creeks, Rivers State, Nigeria

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ABSTRACT

Pollution status of five heavy metals (V, Cd, Pb, Ni, Cu, and Fe) were assessed in Kalabari creeks in the lower reaches of New Calabar River. Sediments, periwinkle (Tympanotonus fuscatus) and water were collected from June 2009 to April 2010 to cover dry and rainy seasons. Sediments were prepared by acid digestion using a mixture of concentrated perchloric acid $HClO_4$, nitric acid (HNO_3) and sulphuric acid H_2SO_4 in the ratio 1:3:1, while periwinkle samples were digested with aqua regia $HN0_3$ and HCl in the ratio of 1:3. Solvent extraction was used to preconcentrate water samples by using ammonium pyrrolidine dithiocarbamate (APDC) and methyl isobutyl ketone MIBK. Bulk Scientific Atomic Absorption Spectrophotometer (A.A.S) model 200A equipped with air-acetylene flame was used for analyses except for vanadium for which nitrous oxide flame was used. The result of ANOVA indicated that the heavy metals were significant at $P<0.05$ in all the samples, while results of t-test at 95% confidence limit indicated no significant difference in mean levels of heavy metals. However, similar trend of levels of heavy metals occurred in water, bottom sediments and periwinkle samples in both seasons. The mean levels of Cd and V were below detection limit (BDL) <0.001 in all the samples investigated. The result revealed that Fe, Ni, Pb and Cu were above FMENV and WHO standards for aquatic organisms. Thus, consumers of periwinkle of the River are at risk of being poisoned by these four heavy metals.

Keywords: Biota, heavy metals, Kalabari creeks, sediments and pollution

INTRODUCTION

The aquatic phase of the environment is an important segment of the total environment. This includes both ground and surface water. This component of the environment plays an important role of accommodating and maintenance of life. Despite this, man continually pollute the water body via various activities such as cultivation of crops, construction of roads, mining, discharged of effluents from domestic and industrial operations which terminate in the surface water through erosion and weathering (Pandey, 2006; Ogri, 2011).

These wastewaters discharged may contain polycyclic aromatic hydrocarbons (PAHs), heavy metals and organic matter. These heavy metals in aquatic environment are partitioned between different medium such as sediment, water, microorganisms and biota. Heavy metals are not toxic in their natural form; rather, they are toxic in ionic form. For instance mercury (Hg) is not toxic as ordinally metal (Hg), rather it is toxic as Hg^+ in methyl mercury ($HgCH_3$) (Laws, 1981). According to Goldwater, (1971) a person could swallow up to half a kilogram or more of metallic (Hg) “with no significant adverse effects”.

According to EPA, (2003) different methods have been used to predict the toxicity of metals. This include, identifying the available fraction of the metal, analyze or calculate the concentration(s) of the bioavailable form(s) in the exposed water and predict the toxicity

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based on an empirical relationship between the biological response and the concentration of the bioavailable forms.

The aquatic organisms such as oysters, scallop and periwinkle are mostly affected by bioaccumulation and magnification of the metal in the food chain due to there feeding habit. According to Manson and Simkiss, (1983) pollutants metal often occur sporadically in trace amount and with different degrees of biological availability. Hence, they are, therefore difficult to detect and quantify by direct analysis of the environment. As a result, a number of organisms have been used as biological indicators to monitor long-term influences within an environment.

Heavy metal pollution differs from other pollutants in that pollutant likes petroleum hydrocarbons are clearly seen in the environment. However, heavy metals may gradually accumulate unnoticed to toxic level and they often occurred in trace amount in water. Sediments serve as sink and metals diffused into the water column when the levels in the water reduced (Horsfall and Spiff, 2002; Okafor and Opuene, 2006; Ayas *et. al.*, 2007). The pollution of river occurred as a result of anthropogenic activities such as mining effluent, industrial effluent, domestic effluents, leaching of metals from garbage, pesticides and urban stormwater runoff.

These pollutants affect the levels of dissolved oxygen DO, pH, dissolved solid, suspended solid, and temperature. According to U.S. EPA (1986) factors which affects the temperature of any water system are; the depth of the water, amount of shade received from shoreline vegetation, the latitude of the waterway, time of the year, volume of the water, temperature of effluents dumped into the water and the colour of the water.

Studies have shown that many water bodies in Nigeria contained variation of levels of heavy metal when compared with develop countries like USA, Germany and Great Britain (Kakulu *et al.*, 1987; Okoye, 1991; Fufeyin, 1994; Idodo-Umeh, 2002). Heavy metals such as copper (Cu), zinc (Zn) and iron (Fe) were reported to be bioaccumulated by aquatic organisms such as oyster and periwinkle, (Lee *et al.*, 1996; Gefford *et. al.*, 2002; Silva *et. al.*, 2003; Blackmore and Wang, 2004; Howard *et. al.*, 2009).

Aquatic organisms such as periwinkles, oysters and scallops are predominant in the brackish water of the Niger Delta and the inhabitants of the area depend on these organisms as source of protein. Thus, it becomes necessary to examine heavy metals in periwinkle and its immediate environment to assess the pollution status in the study area. The aim of this study was to determine heavy metals vanadium (V), nickel (Ni), iron (Fe), lead (Pb), cadmium (Cd) in periwinkle (*Tympanotonus fuscatus),* surface water and sediments.

MATERIALS AND METHODS

Study Area: The Kalabari creeks is located in the lower reaches of New Calabar River (NCR) South-South Nigeria. The New Calabar River is a tributaries of River Niger and River Benue that form a Delta plain in the Niger Delta area. The New Calabar River transverse through five local government areas of Rivers State. The local government areas are Ikwerre, Obio/Akpor, Emohua, Degema, Asari-Toru and finally link to the Atlantic Ocean. The upper reaches of New Calabar River is fresh water while the lower section is brackish water. In the north, the river is bounded by Ikwerre and Obio/Akpor Local Government Area, while in the south is Atlantic Ocean. Some of the communities located in the area are Bakana, Old Bakana, Okporota, Gbukuma, Tombia, Ogbogoro, Sama, Ofouche and Iwofe. The Kalabari creeks are dominated by mangrove trees e.g rhizophora species are found on the inter-tidal area of sediment. The main activities of the inhabitants along the creeks is farming; cultivation of crops and fishing.

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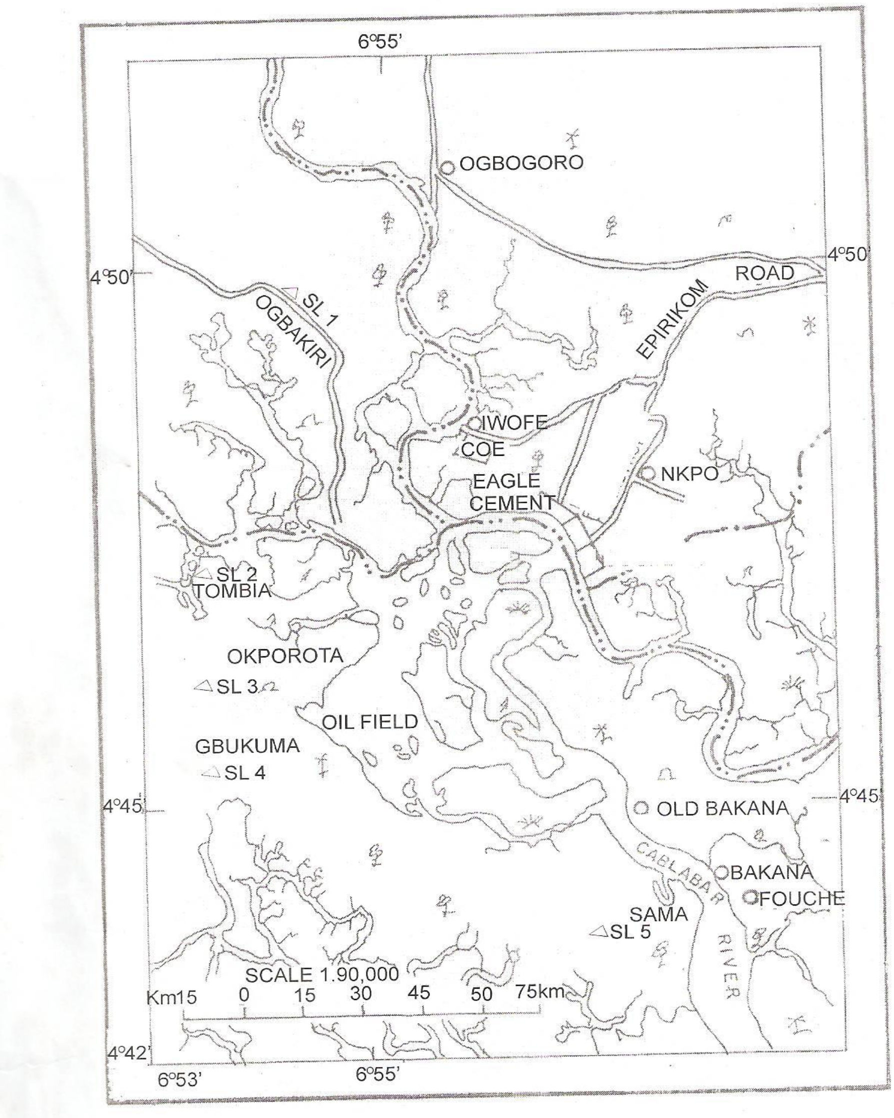

Fig 1: Map showing study and sampling locations.

Sample Collection

Samples were collected from five sites namely; Ogbakiri, Tombia, Okporota, Gbukuma and Sama tagged 1, 2, 3, 4 and respectively during low tide, during this period water ebbs and the surface of the sediments is exposed. The periwinkle samples of various sizes were scattered on the surface of the sediment. Periwinkles of size 3.50-4.0cm were handpicked and washed with water from each site and were placed in a basket labeled according to sites. Surface water samples were collected with nitric acid pre-rinsed 1-litre plastic container from 1cm below the water surface and 5ml concentrated nitric (HNO_3) acid was added immediately in order to reduce chemosorption or hydrolysis of metal ions. Sediments were collected with the aid of a plastic trowel at a depth of about 0-0.5cm and were wrapped in aluminium foil to avoid contamination. Both the surface water and sediments were placed in a cooler packed with ice blocks and was taken to the laboratory.

Sample Preparation

The periwinkle samples were deshelled and the fleshy parts taken and placed in different containers labeled according to sites. The fleshy parts were oven dried at 60^0C for several hours until a constant weight was obtained. On the other hand, sediments were air-dried for three days in the dry season and for one week in the rainy season. Both periwinkle and sediments were ground, sieved and digested according to the method suggested by AOAC, (1995).

According to Golternman *et al.*, (1978), 1g of ammonium pyrrolidine dithiocarbamate (APDC) was dissolved in 100ml of distilled water. This was then transferred to 250ml breaker (with a standard interchangeable stopper) few drops of 2M HNO_3 were added to the breaker until the pH was adjusted to 2.5. 2.5ml of this solution was added to 10ml 2% aqueous solution of methyl isobutyl ketone (MIBK) 4-methly-2-pentanone with stopper and contents of the flask was shaken vigorously for one minute. The two layers were allowed to separate and the organic layer was transfer into 25ml volumetric flask. 5ml of distilled water was used to raise the organic layer into the upper part of the 25ml volumetric flask. The required quantities for analysis were then store in sample bottles pending analysis. The samples were analysed using Atomic Absorption Spectrophotometer model 200A. The data obtained were subjected to one-way analysis of variance (ANOVA) and student t-test at 95% confidence interval.

RESULTS AND DISCUSSION

The mean concentrations of heavy metals obtained from water samples is shown in tables 1 and 2. The result showed that vanadium and cadmium were below detection limit <0.001 in all the months. However, the concentrations of Ni, Cu, Fe, and Pb varied in both sites and months. The mean concentration of all the metals in the three samples were significant at $p<0.05$ levels. The mean levels of all the metals were low in all the months. Nickel recorded the highest mean concentration of 0.79 ± 0.61 mg/L in the month of December and the lowest concentrations of 0.01 ± 0.01 mg/L in the month of October. In December, there was less rainfall and the amount of waste and effluent carried in drains to the creeks from the hinterland were low and water in the creeks becomes highly concentrated. In the Niger Delta area, the month of December and January is usually the peak of dry season, though there may be little amount of rainfall within these two months. The results revealed that Ni recorded the highest concentration in both seasons at site 1, which were 0.025 mg/L and 1.18mg/L respectively. However, the overall mean levels of nickel in both seasons were 0.01 ± 0.01 and 0.43 ± 0.46 mg/L respectively table 1. Table 2 revealed that similar levels of nickel were reported in June, August and October which were months in the dry season.

On the other hand, levels of Ni recorded in the month of December and February is twice the concentration observed in April 2010. Similar trend of Cu, Pb, and Fe were also obtained in the water samples. The mean levels of both Cu and Pb were lower when compared to the mean levels of Fe. The highest mean levels of Cu 0.66 ± 0.54mg/L occurred in the month of February, while the highest levels of Pb 0.13 ± 0.3 mg/L also occurred in February in the dry season. Iron (Fe) recorded the highest mean concentration of the metals examined in water samples. The highest mean concentrations of Fe 3.13 ± 2.73 mg/L was obtained in December Table 2. Iron occurred naturally in the earth crust and is used in most alloys found in our environment such as nail, steel, and in construction companies. A similar result of low levels of heavy metals in water was reported by several researchers in Nigeria (Mombershora *et al.*, 1983; Okoye, 1991; Obire *et al.*, 2003; Emoyan *et al.*, 2006). However the result obtained in this study disagrees with those reported by Wicklund – Glynn, (1991) and Ayas *et al.*, (2007), where higher levels of heavy metals were obtained in water. The mean levels of trace metals in water correlate significantly at 0.01(2-tailed) with levels in periwinkle tissue.

Elevated levels of heavy metals were obtained in sediments when compared to the levels in water. The concentrations of the metals varied from site to sites. Ni recorded the highest mean concentrations at site (1) in the rainy season. However, the overall mean concentrations of Ni 24.43 ± 10.72mg/kg was obtained in the dry season. Copper had the highest mean concentration of 48.3mg/kg at site (2) in the dry season and the overall mean levels of 23.72 ± 13.98mg/kg while Fe had highest mean concentration of 185886mg/kg at site 3 and with the overall mean concentrations of 7777.1±8112.63mg/kg. However, Pb had the highest mean concentration of 5.13mg/kg at site 4 and the overall mean levels of 1.24±1.23mg/kg and 2.04±1.89mg/kg in the rainy and dry seasons respectively, table 3.

The levels of heavy metals Ni, Cu, Fe and Pb in sediments also varied with months. Ni had the highest mean concentration of 25.47±6.31mg/kg in December 2009, while the lowest level occurred in April 2010 which is in the rainy season. Copper also had similar trend of distribution in sediments, the highest mean concentration of 26.84±10.17mg/kg occurred in December, while the lowest occurred in June being the peak of the rainy season. Fe had the highest mean concentration of 9970.1± 97239.78mg/kg in April and the lower levels of 3043.53± 5187.07mg/kg in June.

On the other hand, Pb also had highest concentration of 7.84±1.63mg/kg in April 2010 and lowest mean concentrations of 1.17±1.18 mg/kg in August table 4. The distribution pattern of all the metals corresponded to its natural occurrence in the environment. The results revealed that the levels of heavy metals were higher in the dry season when compared to the levels in the rainy season, tables 3 and 4. The result of one-way analysis of variance (ANOVA) indicated significant difference in the mean concentration of the metals at $p<0.05$. However, the results of t-test indicate no significance in the levels of all the heavy metals in both water and sediments. Similar results of heavy metals level has been reported in water and sediments by Akporhonor *et al., (*2007), Emoyan *et al.*, 2006, Okafor and Opuene (2006), and Ogri *et al.*, (2011) but were lower than those reported by Senthil *et al*., (2008), Vindohini, (2008).

The distribution pattern of heavy metals in periwinkle followed quite a different trend from those in water and sediments table 5. Highest levels of Ni occurred at site (1) in the rainy season when compared to the levels in the dry season. The overall mean levels of Ni in the rainy season was 15.01±10.49mg/kg when compared to 12.55±3.79mg/kg observed in the dry season. The highest mean concentration recorded in site (1) in both seasons implies that site (1) received waste effluents and stormwater runoff from the hinterland before being distributed to other sites.

The overall mean level of copper in the rainy season was 4.54±3.55mg/kg while the overall mean levels of 11.10± 2.76mg/kg was recorded in the dry season. Elevated levels of copper was observed in the dry season in months of December, February and April. This is due to the fact that high temperature results to decreases oxygen levels consequently lead to increased metabolic rate and aquatic organisms take up greater amount of Cu (Ogri *et al*., 2011).

In addition, change in temperature affects the equilibrium between molecular and ionized form of Cu as reported by De Groot *et al*., 1976. The levels of copper obtained in this study agreed with the findings of Akporhonor *et al.*, (2007), Kpee *et al*., (2009). However, the results obtained were lower than those reported by Manson and Simkiss (1983), and Senthil et al, (2008). The distribution pattern of Fe was similar to those of Ni and Cu in periwinkle. Fe recorded the highest mean concentration of 1862.56±122.86mg/kg in periwinkle in February 2010 in the dry season. High temperature in the dry season and elevated total alkalinity level enhance the oxidation of soluble Fe^{2+} to insoluble Fe^{3+} ion state and lower level of these biotic factors at rainy season decreased the bioavailability of Fe to this organism (Nwabueze *et al*., 2011).

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Pb had different trend of bioaccumulation with Fe, lead (Pb) had the highest mean levels of 0.88±0.92mg/kg in August in the rainy season. However, the overall mean concentration of Pb in the rainy season was 0.57±0.78mg/kg and 0.34±0.19mg/kg was obtained in the dry season. The result of analysis of variance (ANOVA) indicate significance difference in the mean levels of Pb with Cu, Fe and Ni in periwinkle, sediment and water at $P<0.05$. The mean levels of heavy metals in periwinkle correlated significantly with the levels in sediments and water. However, the results of t-test indicated no significant difference in the levels of heavy metals in water, sediments and periwinkle, (Table 7). The mean levels of heavy metals obtained in this study revealed that the source of the metals to the environment is mainly anthropogenic. Similar findings were also reported by Okoye, (1991), Okafor and Opuene, (2006) and Akporhonor, *et.al.*, (2007) in Lagos Lagoon and in the Niger Delta area respectively.

CONCLUSION

The mean levels of four heavy metals Fe, Ni, Pb and Cu, in the samples were elevated above FEPA now Federal Ministry of Environment (FMENV) and WHO permissible levels, while vanadium and cadmium were below detection limit (BDL) <0.001 in all the samples analyzed. In periwinkle tissues, the level of heavy metals varied according to sites and months of sampling and they occurred in the order Fe > Ni > Cu > Pb > Cd = V in both the dry and rainy seasons. In surface water samples (SWS), the levels of heavy metals were lower when compared to the mean levels in sediments and periwinkle. The distribution pattern of heavy metals in surface water samples was similar to those recorded in sediments and periwinkle. The mean levels of heavy metals in the samples followed the trend sediments > periwinkle > water. However, fewer situations occurred where the mean levels of heavy metals in periwinkle samples were higher when compared to the mean levels in the sediments. To minimize the pollution of this environment;

(1) The farmers in the area should be educated against the use of chemicals and dynamites as these contain hazardous substance that are dangerous to the aquatic organisms.

(2) The inhabitants of the creeks should be educated against the dumping of domestic wastes and effluents directly into the river.

(3) More studies on the levels of heavy metals and other pollutants should be conducted on periwinkle, water, sediments and other aquatic organisms such as oyster, scallop, mudskipper etc in the New Calabar River system.

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Table 1: Mean levels of heavy metals in water in rainy and dry seasons of NCR system in 2009/2010 in mg/L

Site	*V*		*Ni*		*Cu*		*Fe*		*Pb*		*Cd*	
	RAINY	*DRY*	*RAINY*	*DRY*	*RAINY*	*DRY*	*RAINY*	*DRY*	*RAINY*	*DRY*	*RAINY*	*DRY*
1	BDL	BDL	0.025	1.18	0.001	0.29	5.46	5.80	0.06	0.01	BDL	BDL
2	BDL	BDL	0.002	0.55	0.003	0.76	1.1	2.084	0.003	0.041	BDL	BDL
3	BDL	BDL	0.001	0.09	0.001	0.21	1.18	3.87	0.01	0.03	BDL	BDL
4	BDL	BDL	0.009	0.13	0.003	0.16	0.003	1.03	0.005	0.123	BDL	BDL
5	BDL	BDL	0.014	0.2	0.002	0.003	0.42	0.41	0.02	0.003	BDL	BDL
Overall mean ± Std dev	Nil ± Nil	Nil ± Nil	0.01 ± 0.01	0.43 ± 0.46	0.002 ± 0.001	0.28 ± 0.29	1.63 ± 2.19	2.64 ± 2.20	0.02 ± 0.02	0.04 ± 0.05	Nil ± Nil	Nil ± Nil

Table 2: Mean monthly values of heavy metals in water of NCR in the rainy and dry seasons in mg/L in 2009'2010.

Parameter	*June 2009*	*August 2009*	*Oct. 2009*	*Dec. 2009*	*Feb. 2010*	*April 2010*
V	BDL	BDL	BDL	BDL	BDL	BDL
Ni	0.02±0.01	0.02±0.02	0.01±0.01	0.79±0.61	0.62±0.63	0.32±0.33
Cu	0±0.00	0±0.00	0±0.00	0.21±0.10	0.66±0.54	0.16±0.23
Fe	2.16±2.43	1.56±2.70	1.38±2.17	3.13±2.73	2.83±2.24	0.79±2.05
Pb	0.02±0.04	0.02±0.1	0.07±0.03	0.03±0.4	0.13±0.3	0.06±0.10
Cd	BDL	BDL	BDL	BDL	BDL	BDL

BDL = below detection limit

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Table 3: Mean levels of heavy metals in sediments in rainy and dry seasons of NCR system in 2009/2010 in mg/kg

	V		*Ni*		*Cu*		*Fe*		*Pb*		*Cd*	
Site	*RAINY*	*DRY*	*RAINY*	*DRY*	*RAINY*	*DRY*	*RAINY*	*DRY*	*RAINY*	*DRY*	*RAINY*	*DRY*
1	BDL	BDL	42	25.6	19.3	21.6	559.7	2598	3.06	0.98	BDL	BDL
2	BDL	BDL	32.5	27.4	6.48	48.3	515.9	933.5	1.9	1.22	BDL	BDL
3	BDL	BDL	7.42	15.2	1.43	18	8795	185886	0.77	2.49	BDL	BDL
4	BDL	BDL	12.1	35.62	0.603	15.4	22662	14546	0.29	5.13	BDL	BDL
5	BDL	BDL	9.82	8.33	0.467	15.5	253.1	2222	0.16	0.37	BDL	BDL
Overall mean ± Std dev	Nil ± Nil	Nil ± Nil	20.77 ± 15.5	24.43 ± 10.72	5.66 ± 8.02	23.72 ± 13.98	6557.14 ± 9702	7777.1 ± 8172.63	1.24 ± 1.23	2.04 ± 1.89	Nil ± Nil	Nil ± Nil

Table 4: Mean monthly values of heavy metals in sediments in rainy and dry seasons of NCR system in 2009/2010 in mg/kg.

SITE	*V*		*Ni*		*Cu*		*Fe*		*Pb*		*Cd*	
	RAINY	*DRY*	*RAINY*	*DRY*	*RAINY*	*DRY*	*RAINY*	*DRY*	*RAINY*	*DRY*	*RAINY*	*DRY*
1	BDL	BDL	26.25	16.61	10.40	12.73	449.77	3070.2	0.09	0.307	BDL	BDL
2	BDL	BDL	25.84	11.57	4.55	12.4	346.42	600.33	1.91	0.553	BDL	BDL
3	BDL	BDL	6.59	9.29	3.92	9.72	890.24	1335.31	0.63	0.364	BDL	BDL
4	BDL	BDL	12.26	16.48	2.94	13.74	256.60	1114.62	0.142	0.459	BDL	BDL
5	BDL	BDL	4.133	8.8	0.91	6.89	254.97	1240.63	0.076	0.041	BDL	BDL
Overall mean ± Std dev	Nil ± Nil	Nil ± Nil	15.01 ± 10.49	12.55 ± 3.79	4.54 ± 3.55	11.10 ± 2.76	439.60 ± 264.31	1472.22 ± 937.28	0.57 ± 0.78	0.34 ± 0.19	Nil ± Nil	Nil ± Nil

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Table 5: Mean levels of heavy metals in periwinkle (*Tympanuotonus fuscatus*) in rainy and dry seasons in 2009/2010 in mg/kg

Parameter	*June2009*	*August2009*	*Oct2009*	*Dec2009*	*Feb2010*	*April2010*
V	*BDL*	*BDL*	*BDL*	*BDL*	*BDL*	*BDL*
Ni	11.33±13.14	15.63±13.22	18.08±10.14	15.06±4.87	10.83±7.49	11.76±3.79
Cu	4.23±3.07	4.2±2.81	5.2±8.89	13.1±4.85	10.22±3.75	9.97±4.26
Fe	633.33±37.22	624.92±37.11	264.56±37.12	1573.53±10.46	1862.56±122.86	980.57±15.57
Pb	0.77±1.30	0.89±0.92	0.23±0.22	0.24±0.19	0.27±0.48	0.52±0.34
Cd	BDL	BDL	BDL	BDL	BDL	BDL

Table 6:Mean monthly values of heavy metals in periwinkle (*Tympanotonus fuscatus*) of NCR system 2009/2010 in mg/kg.

		Levene's test for equality of variance		*t-test for quality of means*						
									95% confidence interval of the difference	
		F	*Sig.*	*t*	*df*	*Std. (2-tailed)*	*Mean Difference*	*Std. Error difference*	*Lower*	*Upper*
Score SSS	Equal variances assumed	1.596	.212	-.675	58	.502	-747.69917	1107.0660	-2963.73	1468.334
	Equal variances Not assumed			-675	46.575	.503	-747.69917	1107.0660	-2963.73	1479.968
Score FPS	Equal variances assumed	7.678	.007	-1.248	58	.217	-110.89597	88.83087	-288.710	66.91833
	Equal variances Not assumed			-1.248	35.310	.220	-110.89597	88.83087	-291.176	69.38364
Score SWS	Equal variances assumed	1.149	.288	.971	58	.336	-.29020	.29883	-.88838	.30798
	Equal variances Not assumed			.971	57.072	.336	-.29020	.29883	-.88858	.30818

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Table 7: T-test at 95% confidence levels for surface sediments, periwinkle and surface water samples

				Values obtained in this study		
Parameters /metals	*Water*	*Soils/ sediments*	*Aquatic organism*	*Water*	*Soils/ sediments*	*Aquatic organism*
V	NA	0.1	NA	BDL	BDL	BDL
Cd	0.2-1.8	0.01	0.2-1.8	BDL	BDL	BDL
Pb	1.7	0.2	1.7	0.0-0.13	1.18-2.57	0.27-0.89
Ni	25-150	0.2	NA	0.01-0.79	15.47-25.47	10.22-18.108
Cu	2-4	0.2	2-4	0.00-0.66	5.45-27.35	5.2-9.97
Fe	1.0	5.0	1.0	0.79-3.3	337.24-9970	246.56-1862

Table 8: Comparison of FEPA and WHO permissible levels of heavy metals in sediments water and aquatic organism and this study

Parameter	*June 2009*	*August 2009*	*Oct 2009*	*Dec 2009*	*Feb. 2010*	*April. 2010*
V	*BDL*	*BDL*	*BDL*	*BDL*	*BDL*	*BDL*
Ni	18.56 ± 13.27	24.65 ± 25.98	19.1 ± 8.94	25.47 ± 6.31	24.18 ± 13.24	15.47 ± 9.98
Cu	5.45 ± 7.36	5.9 ± 8.91	5.61 ± 7.84	26.84 ± 10.17	21.11 ± 21.47	27.35 ± 16.06
Fe	3043.53 ± 5187.07	7220.64 ± 106622.2	337.24 ± 88.84	9067.16 ± 84454.35	9163.58 ± 11188.71	9970.1 ± 92739.78
Pb	1.36 ± 1.46	1.17 ± 1.18	1.18 ± 1.07	1.91 ± 1.92	2.57 ± 2.61	7.84 ± 1.63
Cd	BDL	BDL	BDL	BDL	BDL	BDL

NA = Not available, BDL = below detection limit <0.001>

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