ATTENUATION OF HEAVY METALS IN SEDIMENTS OF THE BONNY ESTUARY, NIGERIA: THE ROLE OF OXYGEN

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ABSTRACT

Sediments of the Bonny Estuary, which is repository to pollutants from various industrial and shipment activities in its catchment was investigated for rate of natural attenuation of toxic metals (Cd, Mn, Cr, Pb, Cu, Ni, Fe) under aerobic and anaerobic conditions. Samples were collected with pre-grab sampler from seabed, sieved in the laboratory and monitored under aerobic and anaerobic conditions. After incubation at 18°C, harvests were made on days 1, 14, 28 and 42 and levels of the metals determined spectrophotometrically. Variation plots, ANOVA, Means plots, Student's t-test and Pearson correlation (r) were used to analyze data. Concentration reductions were higher in aerobic condition in the order Cu(15.5%)> Cd(11.4%)> Fe(8.0%)>Mn(5.7%)> Cr(4.4%)> Ni(4.2%)>Pb(3.9%) than in anaerobic condition which was in the order Cu(7.5%)> Cd(7.4%)>Pb(2.4%)>Mn/Cr(1.8% each)> Fe(0.9%)> Ni(0.5%). There was significant heterogeneity in metals reductions (Sig. F=0.000) on all the harvest days, especially of Cu, Pb and Fe at $P<0.05$. Attenuation of Mn also differed markedly between the aerobic and anaerobic conditions (Sig. t=0.005) at the 95% confidence interval. pH influenced the attenuation of Ni (r=0.766) and Fe (r=0.795) ($P<0.05$) as well as Cd (r=0.968), Cr (r=0.861), Pb (r=0.989) and Cu (r=0.950) ($P<0.01$). Results indicate that rate of attenuation was slow and slightly enhanced by oxygenation.

Keywords: Bonny Estuary, sediments, anoxia, heavy metals, attenuation

Academic Discipline and Sub-Disciplines: Environmental Science- Hydrochemistry

Subject Classification: Ecology

Type (Method/Approach): Field and Laboratory experimental procedures

INTRODUCTION

It has been reported that the sediments in many harbours and fjords in regions of the world are polluted by heavy metals and other persistent organic pollutants (POPs). Following this, more than 120 of the polluted sites were investigated between 1993-1996 alone by scientists, and results revealed that about 90 of them were heavily polluted with contaminants such as the polychlorinated biphenyls (PCBs), polynuclear aromatic hydrocarbons (PAHs), tributyl-triphenyl tin compounds, mercury, lead and cadmium [1, 2]. Contaminated sediments are likewise found near population centres and harbours [3-8] such as the Bonny Estuary in the Niger Delta, where there are many sources of pollution of various kinds.

In this estuary where there are industrial installations, including the oil, gas, and ancillary operations located close to the shores, contamination of sediments could be a major environmental problem. Pollutants in sediments can spread to the surrounding water, re-suspend when sediments are disturbed, or be absorbed by benthic organisms through bioaccumulation [9, 10]. Because of certain mechanisms, contaminated sediments may continue to release hazardous chemicals to the surroundings long after the land-based sources of the pollution have been eliminated. As a consequence, contaminated sediments can have serious effects on living organisms [9, 11-15] and the entire ecosystems [16]. POPs and heavy metals may cause higher mortality, reduce growth or disturb reproductive processes in marine organisms. Because many contaminants accumulate in food chains, they can also affect human health through trophic relationships.

The sediment of marine ecosystems are usually depleted of dissolved oxygen; a condition that is generally found in areas that have restricted water exchange. In most cases, oxygen is prevented from reaching the deeper levels by a physical barrier such as mud [8, 17], as well as by a pronounced density stratification, in which, for instance, heavier hyper-saline waters rest at the bottom of a basin. Anoxia, among other effects could tremendously slow down the rate of transformation of pollutants, especially the already persistent ones such as toxic heavy metals from one species to another, including their adsorption to surfaces and particulate matter.

With increasing inputs of pollutants of various nature into the Bonny Estuary by the many oil, gas, shipping, and ancillary companies, there is need to investigate the fate of such POP as toxic metals in a simulated anoxic and oxygenated conditions, as to infer natural removal or at least, attenuation dynamics over time. The concern about heavy metals attenuation in the current study stems from their persistence in the environment; as they are not easily degraded either through biological or chemical means like most other organic pollutants.

MATERIALS AND METHODS

Study Area

Bonny is located between latitudes 4° 22´ and 4° 32´N and longitudes 7° 12´ and 7° 18´E (Figure 1).The weather is hot and humid and subject to heavy rains for about 9 months in the year; with average precipitation of 3800mm, ambient temperature of 33^0C during the day, and humidities of almost 100%. The Bonny Estuary is the main passage for merchant shipping to the inland Port of Onne and for tankers and merchant shipping going to Port Harcourt and the Nigerian National Petroleum Company (NNPC) refined products terminal at Okirika, Rivers State, Nigeria. The estuary receives effluents and other wastes from a number of industries and ancillary companies in the Bonny Island. For example, the Bonny Oil and Gas Terminal (BOGT), the Nigeria Liquefied Natural Gas complex (NLNG), and other service companies are all located proximal to the estuary.

Field Procedures

Triplicate seabed sediment samples were collected from three points located between the NLNG and BOGT facilities in the Estuary with a pre-grab sampler deployed from the deck of a vessel. Successive grabs from about 20 meters depth of the Estuary were emptied into 5 liters containers completely lined internally with aluminum foil to prevent external contamination. Labeled samples were transported to the Laboratory and subsequently preserved in the refrigerator until when needed.
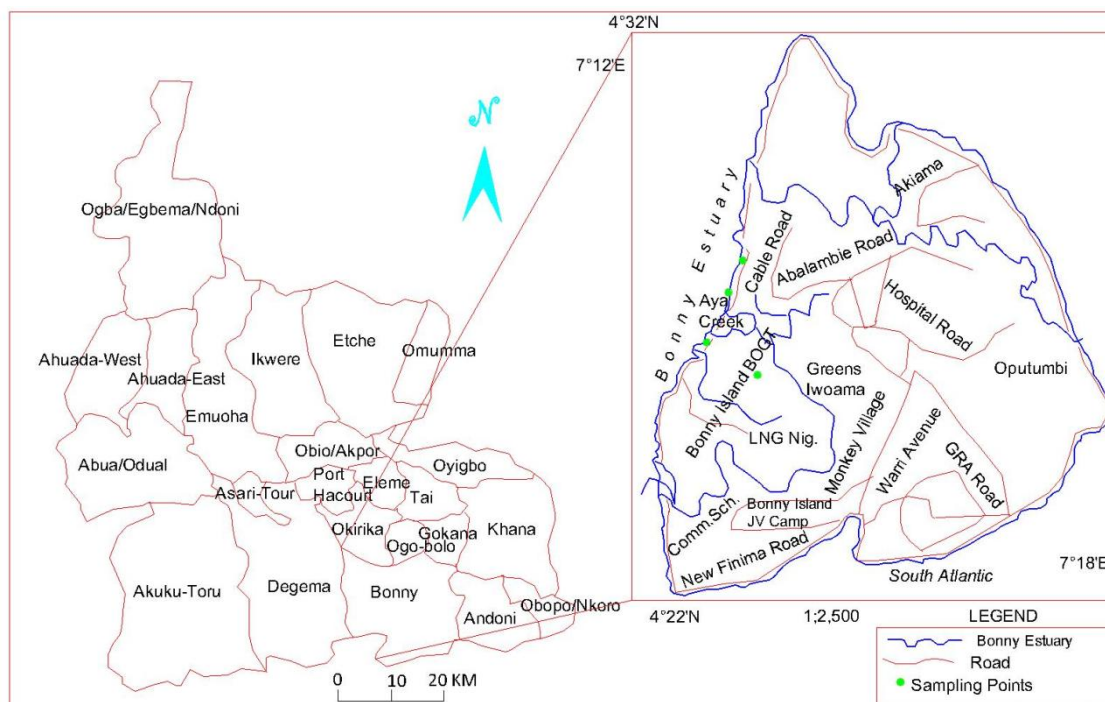
Figure 1. Map of Rivers State showing Bonny Estuary, the study area in the Bonny Local Government Area

Laboratory Procedures

Samples were sieved with habitat water through 2mm, 1mm and 0.5mm mesh size sieves to remove large stones, organic debris and macrobenthos. Sieved sediments were allowed to settle overnight and homogenized by mixing thoroughly with a laboratory desk top mixer for about 10 minutes.

Homogenized sediments were carefully dispensed into three separately labeled glass petri dishes; making sure that the dishes were filled to the top and that there was no air bubbles trapped or overflow of sediment. Three sediment batch samples each, were thus prepared for both anoxic and aerobic experimentations and harvested; making a total of 9 for anoxic and 9 for aerobic setups. The samples for anoxic monitoring were packed in anaerobic jar treated to remove oxygen while those for aerobic monitoring were observed under open laboratory condition. The anaerobic setup was incubated at 18°C and all samples harvested for the determination of toxic metals on days 0, 14, 28, and 42. The pH of sediment samples were also monitored at the same intervals.

Five grams of each sediment sample was weighed into 500mL flask and 10mls of trioxonitrate (V) acid added to digest it by heating in a fume chamber. After digestion, 10mL distilled water was added and the sample filtered. Metal analysis was done with the Spectra AA-600 Varian Atomic Absorption Spectrophotometer by direct aspiration of the digested sample filtrate into an air/acetylene flame generated by a hollow cathode lamp at a specific wavelength peculiar only to the metals programmed for analysis. For every metal investigated, standards and blanks were prepared and used for calibration before samples were aspirated and concentrations at specific absorbance displayed on the system monitor.

Statistical Analysis

Bivariate analyses as provided by the SPSS v.17.0 and the MS Excel 2010 softwares were used. Variation plots were used to represent mean reduction in levels of the metals in sediment samples over the 42 days of experiment. The one-way analysis of variance (ANOVA) was used to test for homogeneity in mean variance of levels of the metals and post-hoc structure of group means detected with means plots at $P<0.05$. The Student's t-test was used to detect marked differences in levels of the metals between the aerobic and anaerobic setups at the 95% confidence interval. The Pearson correlation (r) was used to determine possible significant influences of pH on the metals during attenuation.

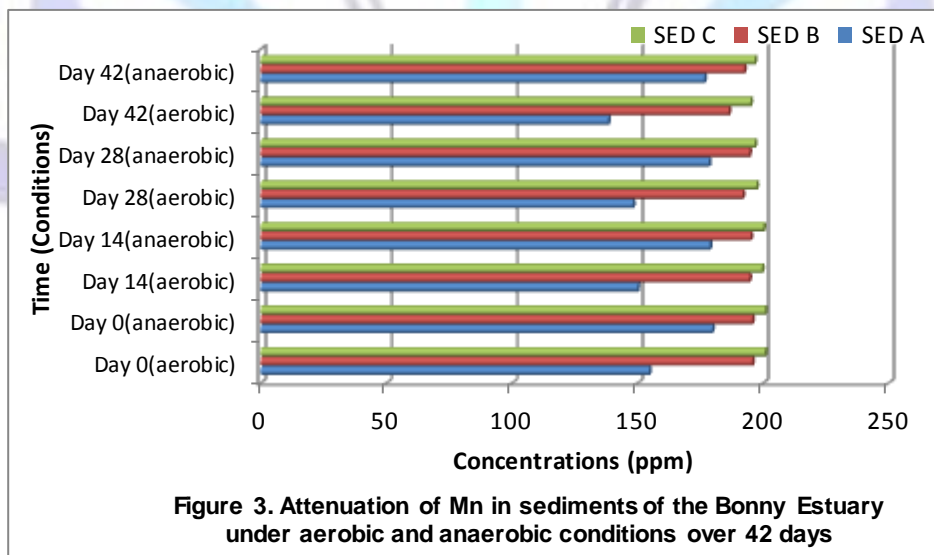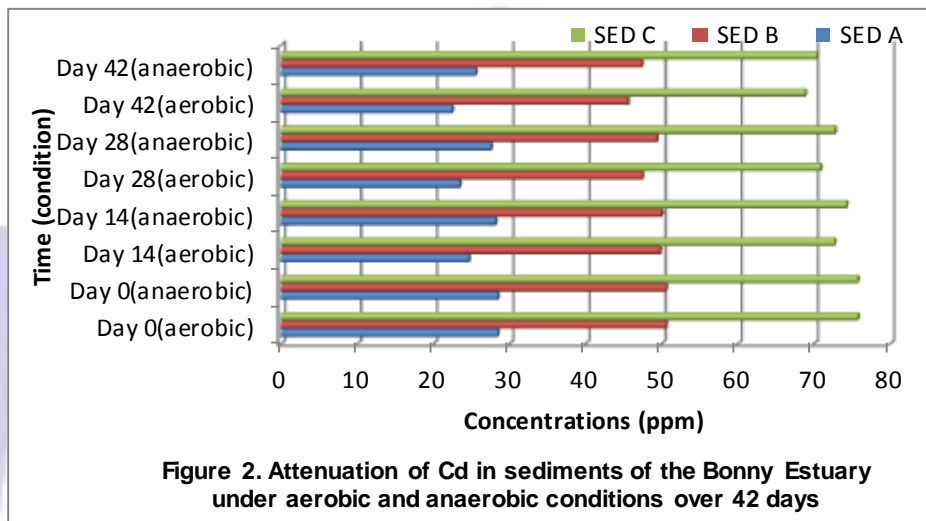## RESULTS

Rate of attenuation of metals in sediments

In sediments A, B and C, Cd attenuated from 28.5, 50.6 and 75.8 ppm at the commencement of experiment through 24.7, 49.8 and 72.75ppm on day 14, 23.5, 47.5 and 70.9 ppm on day 28, to 22.5, 45.6 and 68.9 ppm on day 42 in the aerobic experimental setup respectively (Figure 2). However, it transformed through 28.2, 50.0 and 74.3 ppm on day 14, 27.6, 49.4 and 72.8 ppm on day 28, to 25.6, 47.4 and 70.4 ppm on day 42 in sediments A, B and C of the anaerobic setup.
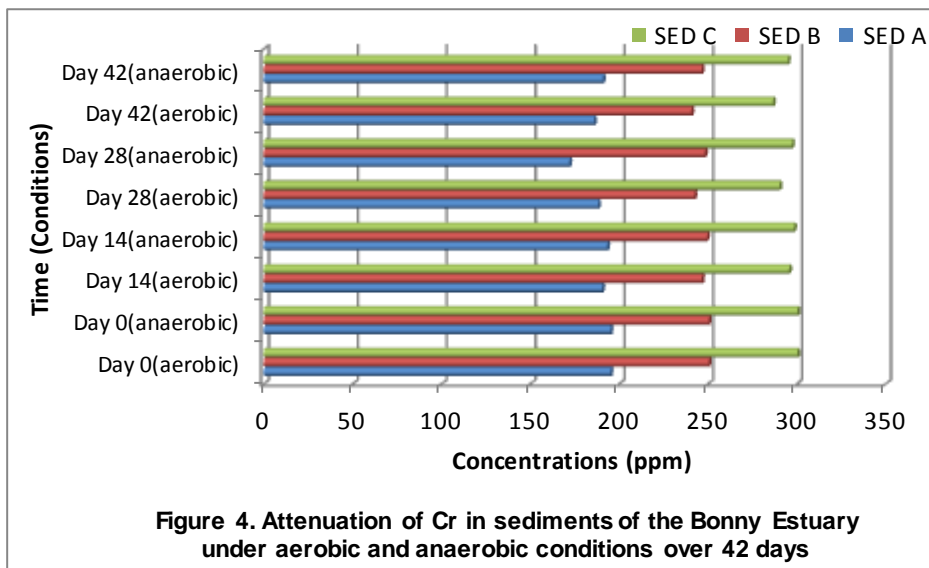
This translates to the percentage reduction in Cd of 11.4 % and 7.4% for aerobic and anaerobic experiments respectively.

For Mn, attenuation in sediments A, B and C was from 154.6, 195.6 and 200.7 ppm at the commencement of the aerobic experimentation through 150.0, 194.5 and 199.5 ppm on day 14, 148.1, 191.8 and 197.3 ppm on day 28 to 138.3, 186.2 and 194.9 ppm on day 42 respectively. This gives a percentage reduction of 5.7%. However, in anaerobic condition, attenuation was from 179.7, 195.6 and 200.7 ppm at the start of experiment, through 178.7, 195.0 and 200.1 ppm on day 14, 178.4, 194.5 and 196.6 ppm on day 28, to 176.5, 192.4 and 196.6 ppm at the end of experiment respectively (Figure 3). This gives a percentage reduction of 1.8%.
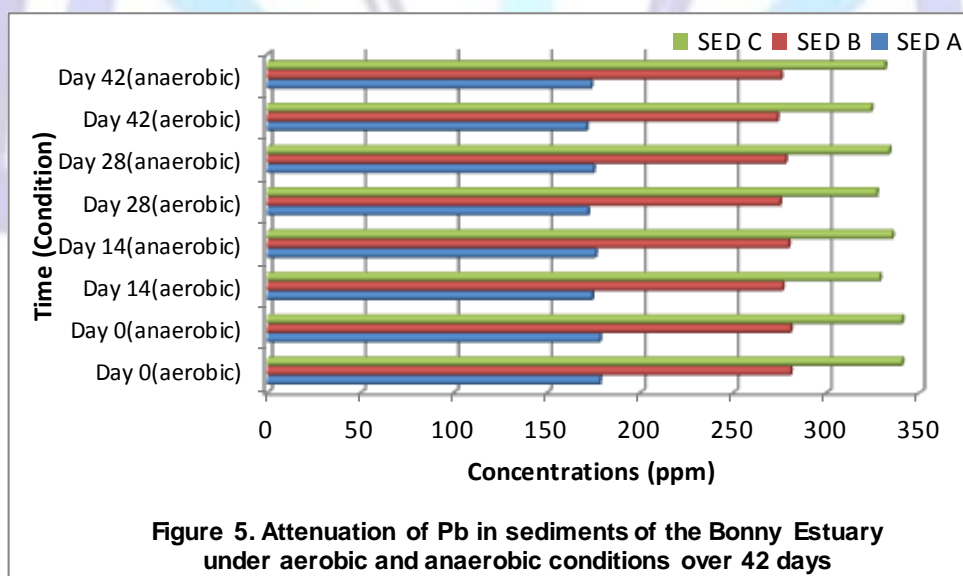
Attenuation of Cr in the sediments samples exposed to aerobic condition was from 195.2, 250.5 and 300.4 ppm on day 0, through 190.5, 246.6 and 295.7 ppm on day 14, 188.2, 242.4 and 290 ppm on day 28, to 185.9, 240.7 and 286.6 ppm on day 42 (Figure 4). This translates to a percentage reduction of 4.4%. Attenuation of Cr in anaerobic setup was through 193.3, 249.5 and 298.5 ppm on day 14, 172.0, 248.3 and 297.3 ppm on day 28 to 190.9, 246.5 and 295.0 ppm on day 42; giving a percentage reduction of 1.8% at the end of experiment.

Figure 2. Attenuation of Cd in sediments of the Bonny Estuary under aerobic and anaerobic conditions over 42 days

Figure 3. Attenuation of Mn in sediments of the Bonny Estuary under aerobic and anaerobic conditions over 42 days

Figure 4. Attenuation of Cr in sediments of the Bonny Estuary under aerobic and anaerobic conditions over 42 days
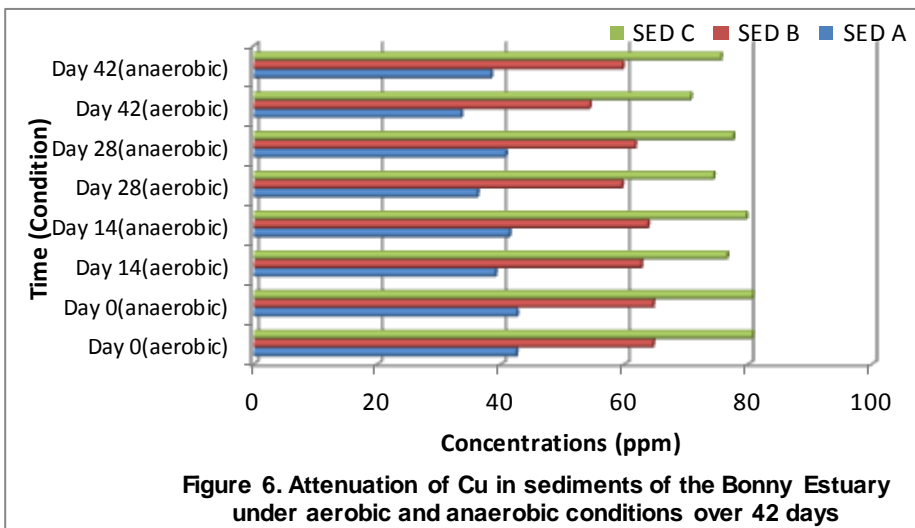
Pb attenuated in the aerobic setup from 178.3, 280.6 and 340.5 ppm at the start of experiment through 174.1, 276.4 and 328.5 ppm on day 14, 172.0, 275.0 and 326.8 ppm on day 28, to 171.2, 273.5 and 324.0 ppm at the end of experiment in sediments A, B and C respectively (Figure 5). In the anaerobic setup however, it proceeded through 176.0, 279.5 and 335.2 ppm on day 14, 174.9, 277.9 and 333.7 ppm on day 28, to 173.6, 275.5 and 331.3 ppm on day 42 in the respective sediments samples; giving a percentage reduction of 2.4%.

In sediments A, B and C, Cu attenuated from 42.4, 64.5 and 80.6 ppm at the commencement of experiment through 39.0, 62.6 and 76.5 ppm on day 14, 36.1, 59.5 and 74.3 ppm on day 28, to 33.5, 54.3 and 70.6 ppm on day 42 in the aerobic experiment respectively; giving a percentage reduction of 15.5%. However, in the anaerobic condition, it attenuated through 41.3, 63.7 and 79.6 ppm on day 14, 40.7, 61.6 and 77.5 ppm on day 28, to 38.3, 59.6 and 75.5 ppm on day 42 respectively (Figure 6). This translates to a percentage reduction of 7.5%.

Figure 5. Attenuation of Pb in sediments of the Bonny Estuary under aerobic and anaerobic conditions over 42 days

Figure 6. Attenuation of Cu in sediments of the Bonny Estuary under aerobic and anaerobic conditions over 42 days
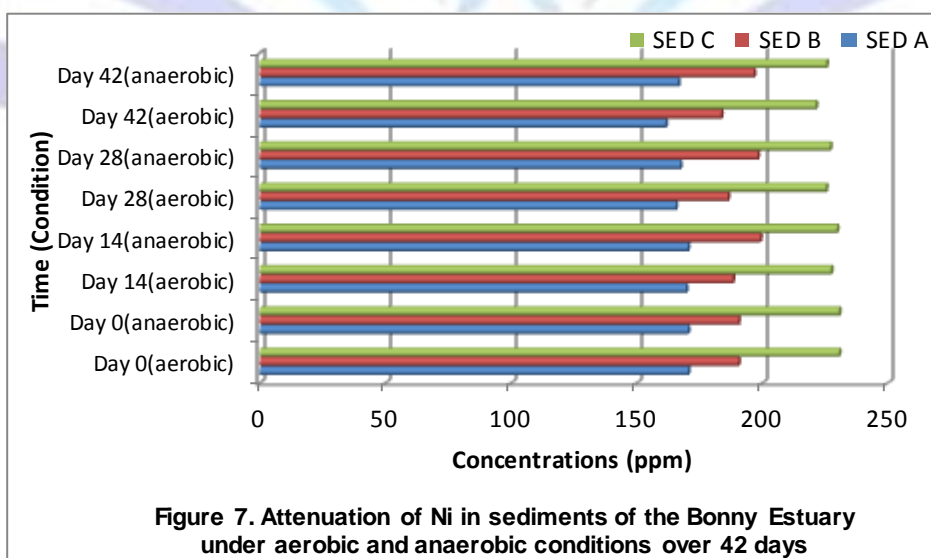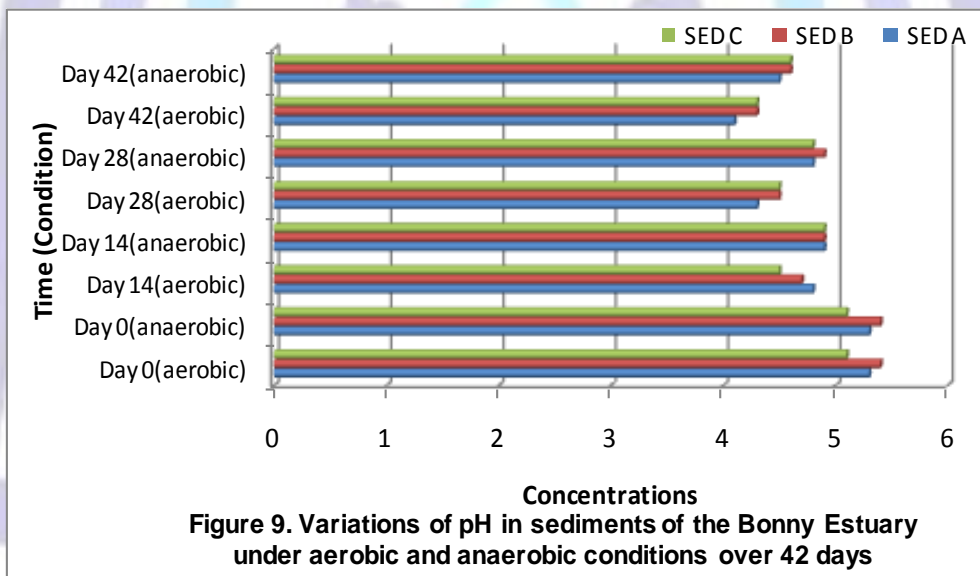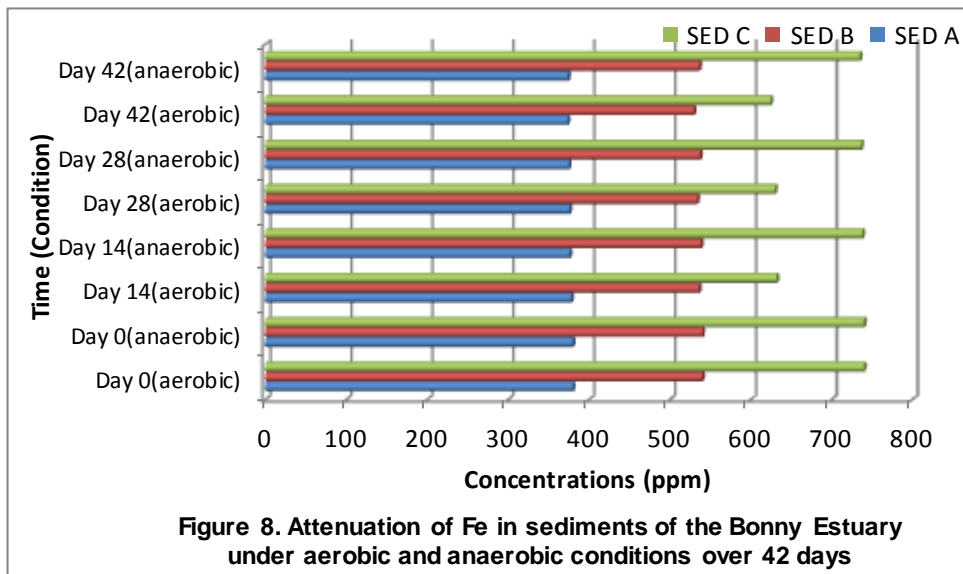
The pattern of attenuation of Ni in aerobic sediments A, B and C was from 170.4, 190.5 and 230.5 ppm on day 0, through 169.6, 188.2 and 227.3 ppm on day 14, 165.5, 186.2 and 225.3 ppm on day 28, to 161.5, 183.6 and 221.3 ppm on day 42 respectively; thus, giving a percentage reduction of 4.2% (Figure 7). However, in the anaerobic setup, it attenuated through 170.3, 199.1 and 229.7 ppm on day 14, 167.3, 198.1 and 227.0 ppm on day 28, to 166.5, 196.6 and 225.5 ppm at the end of the experiment respectively. This gives a percentage reduction of 0.5%.

In sediments A, B and C, Fe attenuated from 380.1, 540.3 and 740.4 ppm at the start of the experiment through 378.4, 536.2 and 632.3 ppm on day 14, 376.0, 534.1 and 630.1 ppm on day 28, to 373.7, 529.6 and 624.7 ppm on day 42. This gives a percentage reduction of 8.0% in the aerobic experimental setup. In the anaerobic setup, attenuation was through 376.1, 538.4 and 738.3 ppm on day 14, 375.3, 537.6 and 736.7 ppm on day 28 to 374.3, 536.4 and 735.1 ppm at the end of experiment respectively (Figure 8). The percentage reduction was 0.9%.

However, pH in the aerobic setup varied from 5.3 in sediment A on day 0, through 4.8 on day 14, 4.3 on day 28, to 4.1 on day 42; 5.4 in sediment B on day 0, through 4.7 on day 14, 4.5 on day 28, to 4.3 on day 42; and 5.1 in sediment C on day 0, through 4.5 on days 14 & 28, to 4.3 on day 42 (Figure 9). In the anaerobic condition, it varied through 4.9 on day 14, 4.8, 4.9 and 4.8 on day 28, to 4.5, 4.6 and 4.6 in sediments A, B and C at the end of experiment respectively.

The test of homogeneity in mean variance over the 42 days experimental period revealed significant difference in rates of attenuation of the metals on days 14, 28 and 42 (Sig. F=0.000 each) at P<0.05. A post-hoc structure of group means that utilized Day 0, the commencement of the experiment as predictor variable revealed that the rate of attenuation of Cr (248.7 ppm), Pb (266.5 ppm) and Fe (553.6 ppm) all in the aerobic experimental setup contributed the heterogeneity most over the 42 days (Figures 10-12).

Figure 7. Attenuation of Ni in sediments of the Bonny Estuary under aerobic and anaerobic conditions over 42 days

Figure 8. Attenuation of Fe in sediments of the Bonny Estuary under aerobic and anaerobic conditions over 42 days

Figure 9. Variations of pH in sediments of the Bonny Estuary under aerobic and anaerobic conditions over 42 days
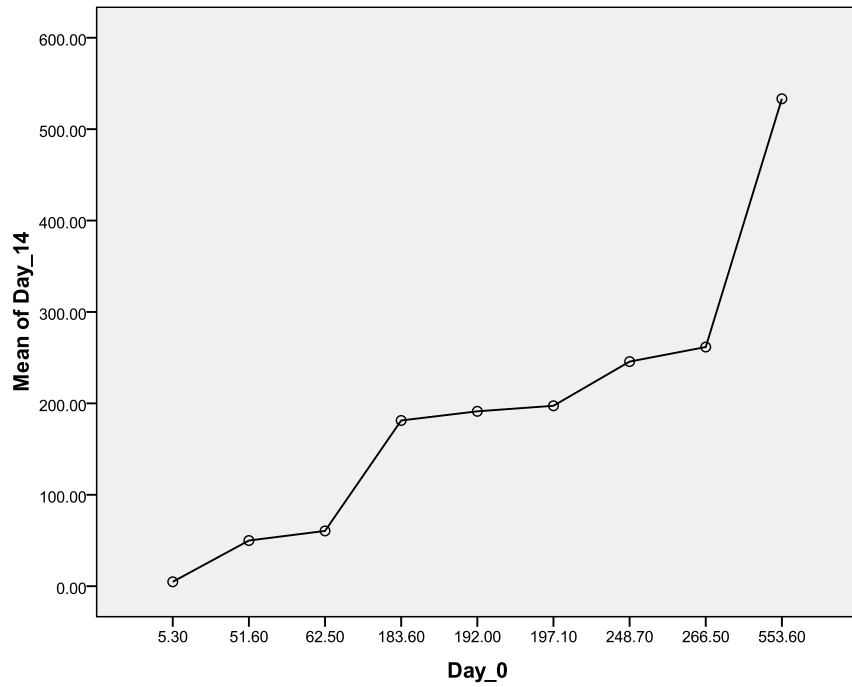
Figure 10. Means plot in attenuation of metals in sediments of the Bonny Estuary on Day 14
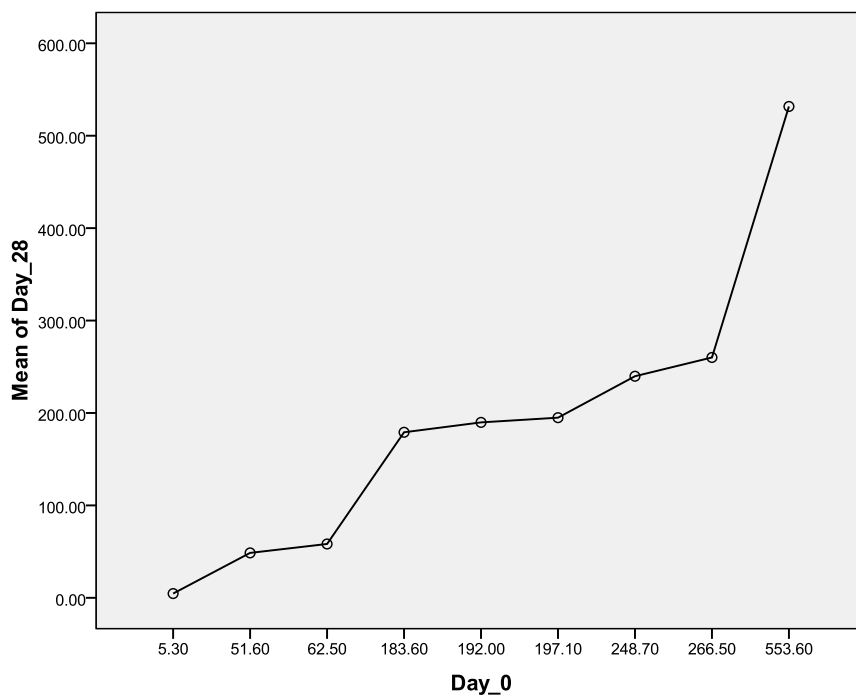

Figure 11. Means plot in attenuation of metals in sediments of the Bonny Estuary on Day 28
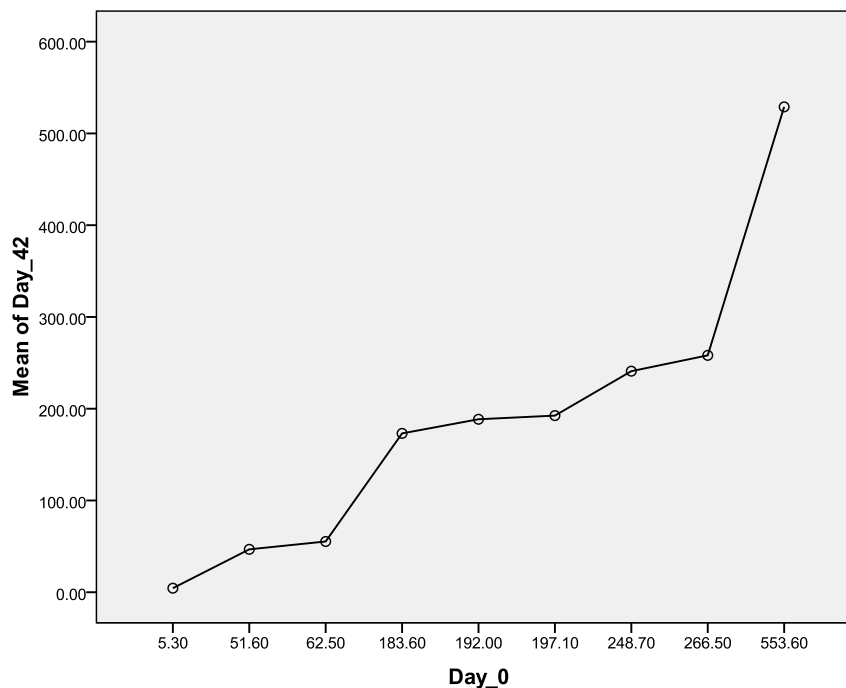
Figure 12. Means plot in attenuation of metals in sediments of the Bonny Estuary on Day 42

Comparison in levels of attenuation of toxic metals in sediments

A pair-wise comparison in levels of attenuation of the metals revealed that in the aerobic experimental condition, mean Cd, Mn, Cr and Pb concentrations were 48.43 (± 1.27), 179.28 (± 2.25), 242.75 (± 2.40) and 260.08 (± 2.26) ppm respectively. Mean Cu, Ni, Fe and pH concentrations were 57.83 (± 2.06) ppm, 193.30 (± 1.79) ppm, 522.98 (± 10.29) ppm and 4.65 (± 0.24) respectively (Table 1).

In the anaerobic condition however, mean concentrations of the toxic metals were as follows: Cd 50.03 (± 0.83), Mn 190.40 (± 0.78), Cr 244.78 (± 2.09), Pb 263.10 (± 1.34), Cu 60.43 (± 1.03), Ni 197.63 (± 0.74), Fe 550.75 (±1.06) ppm. Mean pH was 4.90 (± 0.15).

There was significant co-relationship in the levels of Mn (Sig. r=0.027), Pb (Sig. r=0.031), Cu (Sig. r=0.007) and pH (Sig. r=0.008) between the aerobic and anaerobic experimental conditions at P<0.05. However, the concentrations of Mn differed significantly between the aerobic and anaerobic setups (Sig. t=0.005) at the 95% confidence interval (Table 1).

Relationships between metals and pH during attenuation

Over the attenuation period, pH had significant Pearson correlations with nearly all the metals investigated in the sediments of the Estuary. At P<0.05, pH correlated positively with Ni (r=0.766) and Fe (r=0.795) (Table 2). However, at P<0.01, it correlated positively with Cd (r=0.968), Cr (r=0.861), Pb (r=0.989) and Cu (r=0.950).

Table 1. Pair-wise comparison in metals attenuation in Bonny Estuary using Student's t-test (P<0.05)

Metal (condition)	Mean	SE	r	Sig. r	t	Sig. t
Cd (aerobic)	48.43	1.27	0.943	0.057	-2.836	0.066
Cd (anaerobic)	50.03	0.82				
Mn (aerobic)	179.28	2.25	0.973	0.027	-7.396	0.005*
Mn (anaerobic)	190.40	0.78				
Cr (aerobic)	242.75	2.40	0.731	0.269	-1.210	0.313
Cr (anaerobic)	244.78	2.09				
Pb (aerobic)	260.08	2.26	0.969	0.031	-2.987	0.058
Pb (anaerobic)	263.10	1.34				
Cu (aerobic)	57.83	2.06	0.993	0.007	-2.473	0.090
Cu (anaerobic)	60.43	1.03				
Ni (aerobic)	193.30	1.79	0.505	0.495	-2.779	0.069
Ni (anaerobic)	197.63	0.74				
Fe (aerobic)	522.98	10.29	0.945	0.055	-2.988	0.058
Fe (anaerobic)	550.75	1.06				
pH (aerobic)	4.65	0.24	0.992	0.008	-2.611	0.080
pH (anaerobic)	4.90	0.15				

SE=standard error of mean, *=significant at P<0.05

Table 2. Correlation (r) matrix between the metals and pH in sediments of the Bonny Estuary over 42 days

	Cd	Mn	Cr	Pb	Cu	Ni	Fe
pH	0.968**	0.682	0.861**	0.989**	0.950**	0.766*	0.795*

*=significant at P<0.05, **=significant at P<0.01

DISCUSSION

The current study clearly revealed that the toxic metals investigated were recalcitrant to degradation, especially in anoxic conditions of sediments in the wild marine environment. Elsewhere, the persistence of heavy metals has already been noted by Ogbuagu *et al.* [10] and Victor *et al.* [18], among many others. They can only be transformed from one ionic form to another. Microorganisms could sparingly degrade them, only when enhanced by stimulations or other augmentation techniques. The slight reductions in metals concentrations were most likely due to adsorption to surfaces and particulate matter. The most reduced metal was Cu and the least Pb in oxygenated condition, and Cu and Ni, respectively in anoxic condition. In the aerobic condition, the order of rate of reduction was Cu>Cd>Fe>Mn>Cr>Ni>Pb and in the anaerobic condition, the order was Cu>Cd>Pb>Mn/Cr>Fe>Ni. Generally, the metals reduced in concentrations more in the aerobic than anoxic setups. This underpins the role of oxygen in the enhancement of biodegradation and transformation processes of metals in aquatic ecosystems.

The observed statistical heterogeneity in rates of attenuation of the metals clearly indicates that concentrations varied markedly during the period. This implies that duration or time is a determinant factor in the transformation or attenuation of pollutants in at least, the aquatic environment. The most implicated metals that were influenced by time factor are Cu, Pb and Fe, whose levels in sediments of the Estuary may have mostly been elevated by allochthonous industrial inputs in the catchments of the estuary. Industrial inputs from the oil and gas, as well as ancillary companies operating in the Bonny Island may have left this footprint of elevation in pollutants loading. Other activities that may have contributed these toxic pollutants includes shipping and transport activities which are rife in the Estuary.

Trend in attenuation of Mn differed significantly between the aerobic and anaerobic setups. This again underscores the role of oxygen in sediment pollutants detoxification and purification.

The observed positive correlations between the metals and pH collaborates seeming acidification that was associated with decreases in metals concentrations in the sediments over the 42 days period. Accordingly, increasing acidification was accompanied by lower concentrations of the metals during attenuation.

Summary

In the aerobic condition, the most attenuated toxic metal was Cu and the least Pb, and in the anoxic condition, the most attenuated was also Cu but the least, Ni. Reduction in concentrations was higher in the aerobic than the anaerobic condition. Attenuation differed markedly over the experimental period, especially in Cu, Pb and Fe. pH had significant positive influences on the metals during attenuation.

Conclusion

This study revealed that metal species in marine sediments were persistent under natural degradation process, and that oxygen slightly enhanced their attenuations in the environment.

ACKNOWLEDGMENTS

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Corresponding Author's brief biography

Dike Henry Ogbuagu, an ecologist and environmental biologist, holds a doctoral degree in Hydrobiology from the University of Ibadan, Nigeria. He is also a lecturer with the Department of Environmental Technology, Federal University of Technology, Owerri, Nigeria. His current research interest is ecotoxicology and he has several publications with national and international peer-reviewed journals.