SPATIAL DISTRIBUTION OF TRACE ELEMENT OF ALA RIVER'S SEDIMENTS, AKURE, SOUTHWESTERN NIGERIA

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ABSTRACT

Thirty six river sediment samples were collected from Ala River, Akure, and were analysed by ICP-MS for 14 chemical elements including Cu, Pb, Zn, Ni, Co, Mn, As, U, Th, Sr, Cd, V, La, and Cr. The concentration levels of the metals ranged from 0.26 µg/g in Cd to 3719.00 µg/g in Mn. Strong variations in sediment geochemistry were observed from one sample location to the other. Several hot-spot areas were located close to areas where activities such as vehicle related workshops, road junctions, municipal waste, abattoir and farm gardens were identified. The concentration levels of Pb (19.13-164.54 µg/g), Mn (354-3719 µg/g) and Zn (49.30-749.50 µg/g) were of environmental concern because of their relatively high concentrations in the sediments, at some locations, when compared with the average earth crust background limit. Most of the other elements did not show much contamination load as compared with the average earth crust background limit. The geo-accumulation index (I-geo) values showed that the sediments were contaminated with Pb, Zn and Cu. Metals such as Pb, Zn, Cu, Cr, Mn, Ni, As, Cd and V were positively correlated with each other (r = 0.51-0.95), which indicated analogous sources of geochemistry characteristics. The study showed that anthropogenic activities greatly contributed to enrichment of trace elements in Ala River sediments.

Keywords: Ala River, Sediment, Geo-accumulation index, spatial variation, anthropogenic

INTRODUCTION

Stream Sediments are classic materials that are derived from the parent bed rock, as well as sand and deposited particles. Urban river sediment geochemistry is receiving increasing attention due to the potential long-term toxicity of some heavy metal pollutants resulting from urban activities (Nyarko *et al.*, 2006; Ajmone-Marsan *et al.*, 2008). Sediment contamination and subsequent pollution resulting in continuous enrichment of heavy metals such as Pb, Zn, Mn, Cu and Ni, is of great concern worldwide (Kabata-Pendias and Mukherjee, 2007). Elevated concentration of some of these metals in stream sediment poses great threat to public health due to the possible chain of leaching into the entire biota consumables which include leaching into groundwater, entering the food chain through plant uptake, direct human and other animals' consumption via surface water or the hand-to-mouth pathway.

The toxicological risks of consumption of water polluted by sediment loaded with heavy-metals are especially high for children due to their physiological characteristics (Ajmone-Marsan *et al.*, 2008; Carr *et al.*, 2008; Morton-Bermea *et al.*, 2009). Akure metropolis is one of the many cities in Nigeria that have continued to witness tremendous growth in human population. Market places, high waste generation, increased vehicular activities, raw material processing industries, saw mill, abattoir, non seasonal farming along river/stream channels with fertilizer input, and waste metal recycling company are some of the activities resulting in generation and accumulation of waste. These wastes eventually find their ways into most of the drainages because of obvious non- regulation of waste disposal by government authorities. There are many streams that drain Akure metropolis, but Ala River is the main river that drains the town. This makes the river very important for geochemical assessment. There is no known published work that has been carried out on the investigation concerning metal pollution status of this river.

The city lies within Longitudes 007° 14^{l} to 007° 19^{l} and Latitudes 05° 08^{l} to 05°. 15^{l} (Fig 1). It is connected by roads linking Ondo town in the south, Owo in the east, Ado-Ekiti in the north and Ilesa town in the west, with an average distance of between 45 km to 60 km to all these cities. The city cover's an average land area of 72 km^{2}. The Ala

River is about 13 km long within the city, and drains into Ogbese River in the eastern part of the metropolis. The city is made up of communities like the Federal University of Technology, Akure, Aule, Oba Adesida Road, Ijapo, Sabo, Isikan, Otutubiosun, and many other communities that developed to form the city.

The area falls within the tropical rain forest of Southwestern Nigeria, where there exists two major seasons; wet and dry seasons. The wet season usually spans through the months of April to October, while the dry season is usually between November and March. The average annual rainfall is about 3000 mm (Iloeje, 1980). The area falls within the Crystalline Basement Complex of Southwestern Nigeria (Rahaman, 1989). The local rock units consist of migmatite-gneiss, quatzites, Older granites and charnockites (Owoyemi, 1996) (Fig. 2). The Older granites and charnockites exist together as single body in all the outcrops, while the migmatite-gneisses occur separately (Adeyemo and Omosuyi, 2012) low lying outcrops in some areas, while in some other areas, they occur as massive and extensive outcrops. The mineralogy consists of biotite, feldspar, quartz and muscovite minerals.

The aim of this study was to investigate the spatial variation of urban geochemistry with a focus on heavy metal concentrations in the sediments of Ala River. It has been established that the pathway between pollutants and receptors is very short (Tijani *et al*., 2004, Dao *et al*., 2010). Such study therefore, will enable the establishment of the pollution status of the Ala River sediments.

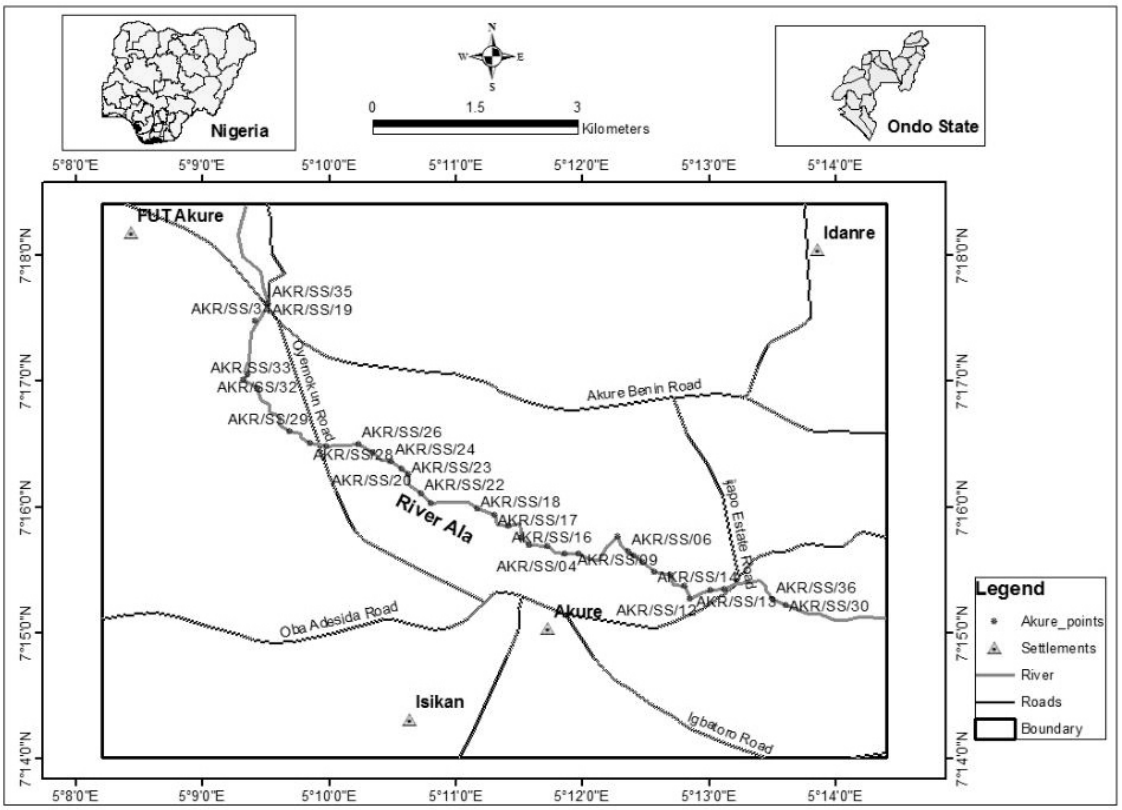


Fig. 1: Sampling Locations of the Study Area (Modified from Sheet 264, Federal Survey, Nigeria, 1966)

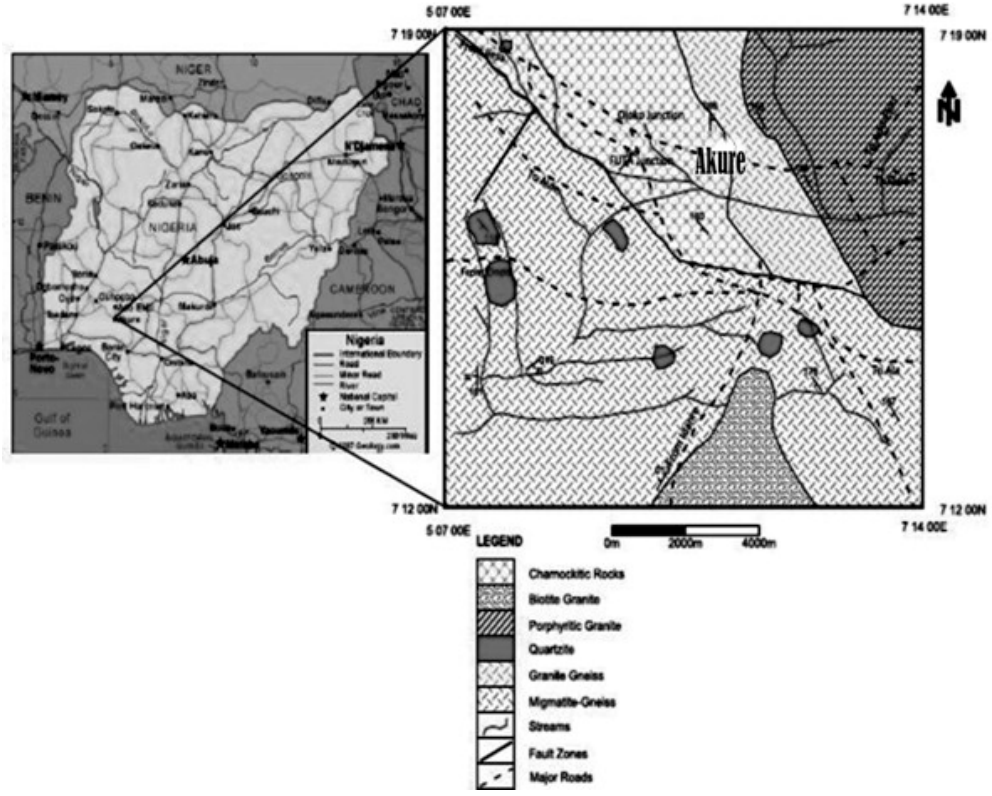


Fig. 2. Simplified Geological Map of Akure Metropolis (Modified after Owoyemi, 1996),

MATERIALS AND METHODS

Sampling and Chemical Analyses

An intensive investigation of Ala River sediments was carried out during the period of February 2014 (dry season) and a total of 36 sediment samples were collected along the river bed at an interval of between 200 m – 300 m per location (Fig. 1) using a stainless steel pan. The samples were emptied into a plastic bucket in the river. Each sample is a composite sample, consisting of 4-5 sub-samples taken from 1-2 m radius within a chosen location. The approximate mass of each primary sample was 300-400 grams, collected from a depth of 0-10 cm right at the center of the river. Published works have shown that such depth in the river bed serves as the main receptor and sink for metal enrichment (Olatunji and Abimbola, 2010) and because the depth range is the most obvious fingerprint of environmental contamination, (Dao et al., 2010) in sediments.

Sediment samples collected were sieved using the wet sieve method from the plastic bucket using < 65 μm stainless steel mesh to remove stones and plant debris and further mixed thoroughly using the water from the river in other to obtain a representative sample. The mixed sediment samples were allowed to settle and the liquid mixture were further sieved with < 53 μm mesh into another bucket and the clay fractions were stored in a 150 cl plastic bottle and taken to the laboratory. To avoid any form of cross contamination, all devices were thoroughly cleaned using the stream water after each sample was processed. Again, because the samples were collected wet, the samples were allowed to settle down in the stored plastic bottle for some hours, and the clay fraction was allowed to get dried under room temperature. Thereafter, the dried samples were now pulverized using an agate mortar and pestle and further sieved with <53 μm for subsequent sample digestion and chemical analyses. The samples were digested using aqua-regia digestion (0.5 ml H_2O_2), 6 ml concentrated HNO_3 and 1.8 ml HCl) for 2 hours at 95°C. The samples were cooled and then diluted to 10 ml with deionized water. The digested samples were analyzed to determine the constituents of the elements using Inductively Coupled Plasma-Mass Spectroscopy (ICP-MS) at the Geochemical Laboratory of ACME laboratory Limited, Vancouver, Canada.

Subsequently, the analytical results of fourteen trace elements, namely; Cu, Pb, Zn, Ni, Co, Mn, As, U, Th, Sr, Cd, V, La, and Cr, were selected for qualitative treatment and quantitative statistical analysis. Also, for risk assessment, the results were compared with published works on baseline mean concentrations of sediments from pristine areas and similar geologic environment and the pollution index factors were analyzed using conventional statistical interpolations for each sample location.

RESULTS AND DISCUSSION

The descriptive statistic summary result of trace element concentration (μg/g) of Ala River sediments is presented in Table 1 and the box plot of the total concentrations of the elements is presented in Figure 3 in terms of percentile distribution representing the mean, median, 25th and 75th percentiles. From the results, it was observed that there is significant heterogeneity in the concentrations of the elements being studied, with the obvious spatial variation of concentrations from one sample location to another.

In the river sediments Mn was found to exhibit the highest concentration in (μg/g) range = 354-3719, mean ± standard deviation= 1221.31 ± 765.62), followed by Zn, (49.30-749.50, mean 257.08 ± 197.17), Pb (19.13-164.54, mean 56.67 ± 41.51), V(29.00-172.00, mean 79.77 ± 26.46), Sr (14.30-91.40, mean 46.90 ± 46.90), Cr (19.90-59.60, mean 41.77 ± 9.21), Cu (8.00-112.59, mean 34.75 ± 20.21), La (32.70-705.00, mean 48.63 ± 9.35), Co (9.70-57.00, mean 26.18 ± 10.49) and Ni (5.50-32.70, mean 14.45 ± 4.63). The mean concentration was below 0.60 μg/g for As, below 12.06 μg/g for Th, 1.55 μg/g for U and below 0.26 μg/g for Cd.

Higher concentrations of Mn , were observed at AKR/SS/24 (3719 μg/g). Other locations with relatively high concentrations above 1000 μg/g included AKR/SS/(8, 10, 13,14, 17,18,19,21,27,26,31,32 and 34) with concentrations of (1052, 1291, 2287, 1081, 1468, 1050, 2447, 1608, 1765, 1633, 2158, 3020 and 1853 μg/g), respectively. The location with the

lowest concentration of Mn was AKR/SS/12 (354 μg/g). The varying dispersion of Mn concentrations from one location to another, indicated a relatively high concentration of Mn, and this could have been influenced by varying anthropogenic factors such as engineering tools and vehicle repair activities.

Zinc spatial distribution showed that location AKR/SS/2 with concentration of 749.5 μg/g, had the highest concentration of Zn. Other locations with relatively high concentrations included AKR/SS/3 (641 μg/g), AKR/SS/4 (522.6 μg/g) and AKR/SS/16 (717.6 μg/g). Locations that were found to have relatively moderate Zn concentrations included AKR/SS/13 (307.4 μg/g), AKR/SS/14 (320.9 μg/g) and AKR/SS/15 (441.5 μg/g). The lowest concentration of Zn was found in AKR/SS/12 (49.3 μg/g). Comparing the mean concentration of Zn in the river sediments with published baseline rock values, (Table 2) of Earth crust (70 ppm), Ultramafic (50ppm), Basalt (100 ppm), Granodiorite (60 ppm) and Granite (40 μg/g) (Levinson, 1974), it was found that the mean concentration of the Zn (257.08 ppm), in the study area was higher, which suggested that the element might have been enriched by anthropogenic sources.

Relatively high concentrations of Pb were observed at locations AKR/SS/2 (160.1 μg/g), AKR/SS/3 (124.01 μg/g), AKR/SS/4 (164.54 μg/g) AKR/SS/8 (112.75 μg/g) and AKR/SS/9 (110.94 μg/g). Other locations with relatively moderate to low concentrations were generally below 100 ppm in Pb concentration. The lowest concentration of Pb (19.13 μg/g).was found at location AKR/SS/12. Most of the elements followed the same trend on elemental concentrations, suggesting that the enrichment of these elements in the media analysed might have been influenced by similar sources. The other elements i.e V, Cr, Ni, Co, La, Th, U, As, Sr, and Cd, also showed spatial distributions that aligned with the effects of anthropogenic sources (areas close to waste dump, vehicular activities, farm lands, domestic emissions, wear and tear of vulcanized rubber tires) for some locations. Areas with high elemental concentration followed the same trend for most of the metals observed. The relative pervasive general metal enrichment in almost all the locations for Pb, Mn and Zn suggested high effects of urban activities. Severe contamination appeared to be of the same trend in most localities for Mn, Zn and Pb, suggesting noticeable pollution in Ala stream sediments, because of the high influx of waste draining into the river.

Table1: Descriptive Statistics of Trace Element Concentration (μg/g) in River Sediment of the Study Area

Element	N	Minimum	Maximum	Mean	Std. Deviation
Cu	36	8.00	112.59	34.75	20.21
Pb	36	19.13	164.54	56.67	41.51
Zn	36	49.30	749.50	257.08	197.17
Ni	36	5.50	32.70	14.45	4.63
Co	36	9.70	57.00	26.18	10.49
Mn	36	354.00	3719.00	1221.31	765.62
As	36	0.10	1.50	0.60	0.52
U	36	1.14	2.32	1.55	0.23
Th	36	7.80	17.00	12.06	2.20
Sr	36	14.30	91.40	46.90	19.18
Cd	36	0.04	0.84	0.26	0.20
V	36	29.00	172.00	79.77	26.46
La	36	32.70	70.50	48.63	9.35
Cr	36	19.90	59.60	41.77	9.21

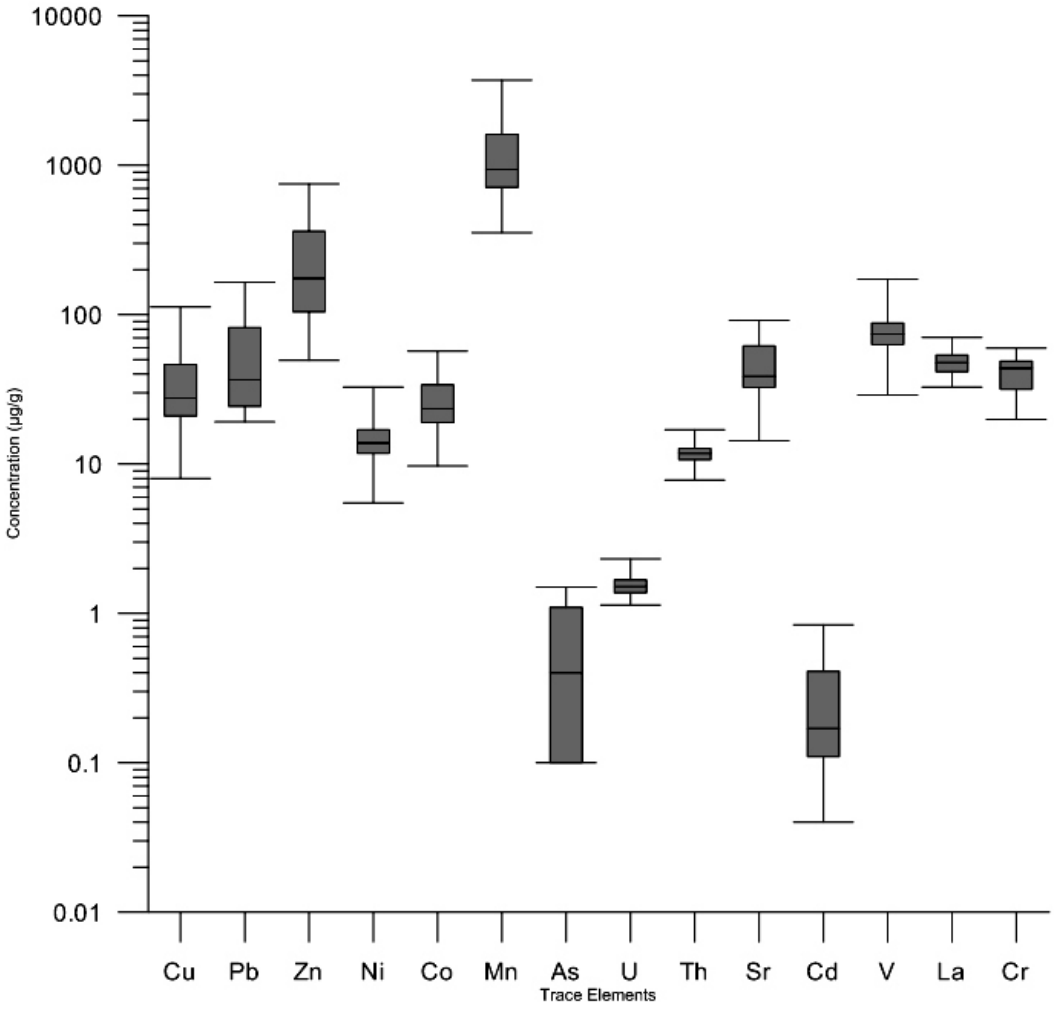


Fig. 3: Box Plots of the Total Concentrations (µg/g) of Trace Elements in River Sediments (The central box represent 50% of the distribution, between the 25th and 75th percentiles. The line bisecting the central box is the median: the whiskers extend to the Max and Min of the distribution).

Table 2. Comparing the Mean of Elemental Concentration of this Study and Average Abundance of fourteen Trace Elements in the Earth Crust and Various Rocks (ppm) (after Levinson (1974), Adepoju and Adekoya, 2011)

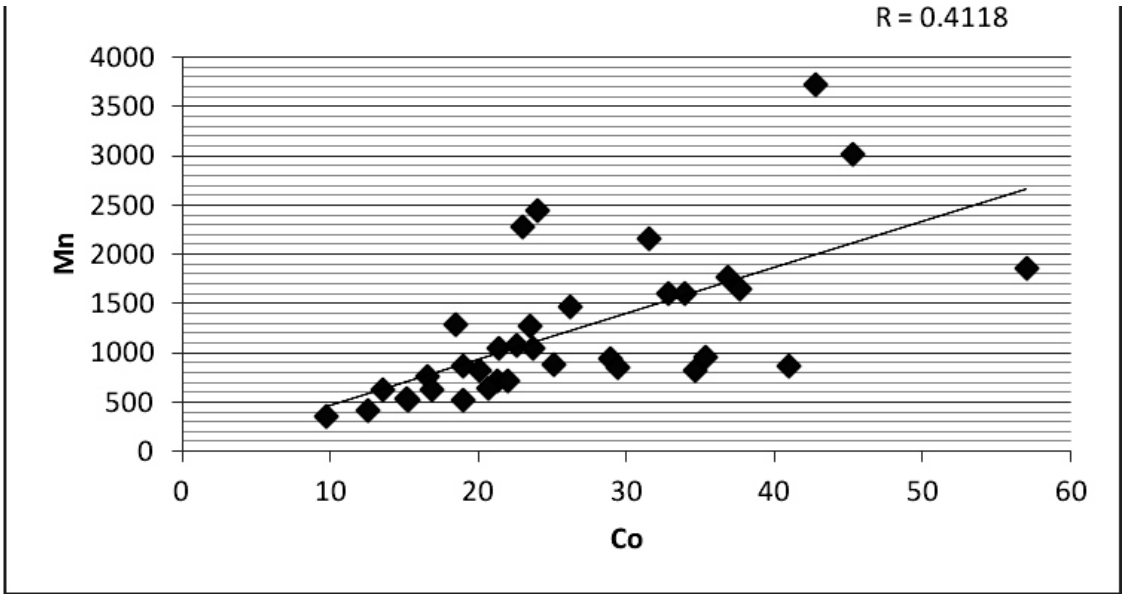
Element	Earth crust	Ultramafic	Basalt	Granodiorite	Granite	Mean of present study
Cu	55	10	100	30	10	34.75
Pb	12.5	0.1	5	-	20	56.67
Zn	70	50	100	60	40	257.08
Co	25	150	50	10	1	26.18
As	1.8	1	2	2	1.5	0.60
Ni	75	2000	150	20	0.5	14.45
Cr	100	2000	200	20	4	41.77
La	30	3.3	10.5	36	25	48.63
U	2.7	0.001	0.6	3	4.8	0.23
Th	10	0.003	2.2	10	17	12.06
Sr	375	1	465	450	285	46.90
V	135	50	250	100	20	79.77

Statistical Analyses

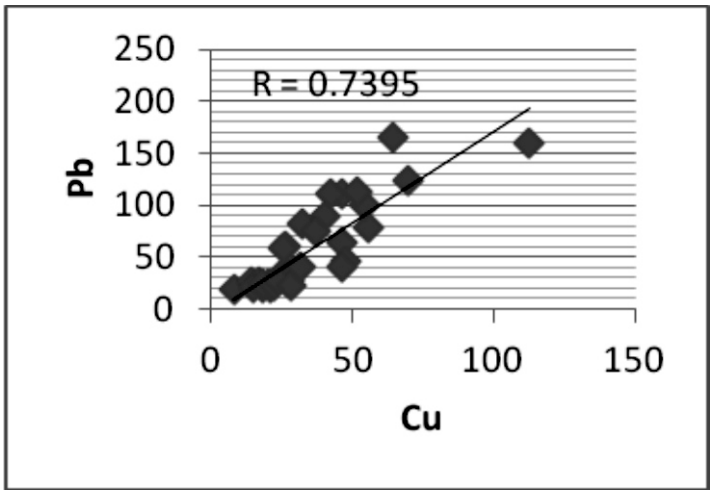
The determined geochemical results were further subjected to statistical interpolation to ascertain the relationship between the various elements. Both Pearson Correlation (PC) and Principal Component Analysis (PCA) were carried out using Origin and SPSS software packages. The Pearson Correlation showed a wide variation in the inter elemental relationship both for the Scatter Plot and Correlation Coefficients of the trace elements [Figure 4 (a-e) and Table 3]. For the elemental relationship, there is a fair coefficient of determination (R) between Mn and Co (R = 0.4118) and between Zn and Cr (R = 0.4623), while there is strong correlation between Cu and Pb (R = 0.7395), Cd Vs As (R = 0.7232) and Zn and Pb (R = 0.6628). From the affinity level of the elements, it could be that these elements were influenced by analogous sources of enrichment across the sediments studied. The correlation matrix for the data obtained (Table 3) shows a wide variation in the correlation coefficient (r) between element pairs. For example, the r values ranged from −0.01 between Th and Co to 0.95 between Cd and Zn. Fairly strong to very strong positive correlations occured among Pb, Cu, Zn, Ni, Cd, As and Sr. Considerably high correlations were found between Pb and Cu (r = 0.86), Zn and Cu (r = 0.88), Zn and Pb (r = 0.81), Cd and Zn (r = 0.95), and V and Ni (r = 0.92). Other elements also showed fair to strong correlation as summarized in (Table 3).

In a similar vein, the results obtained from the two PCA and three component plot, (Table 4, Figure 5) further clarified elemental association in four and three component factors of association, respectively. Factor analyses revealed four groups of trace elements (Table 4); the first group is made up of Cu, Pb, Zn, As, Sr, Cd, and Cr accounting for 49.32% total variance of eigen value. These elements as also found in the correlation values (Table 3), might have been enriched by same sources, such as from domestic waste, effuents and vehicular wastes. Though some of the elements like As and Cd are relatively low in concentration, their presence nevertheless as earlier stated, could be tied to close or similar sources with the other elements. The second group is made up of Co, Mn, Ni and V, accounting for approximately 22.16% of the total variance. These elements might have been enriched in the sediments by activities relating to similar source draining into the stream. Action of weathering and transport of the underlining rock might have added to the enrichment sources. But for the relatively high concentration of Mn, there also may be possibility of anthropogenic source from waste leachate, such as alloys for corrosion resistant equipment and batteries. The third group consists of U, Th and La, which accounted for 12.94% of the total variance. These three elements might have been enriched by activities relating to soil weathering and leachate from automobile activities and other urban consumables like additives in chemical products. Mn occurred in association with Ni, Co and V, with a total eugen value of 5.08%. This occurrence may have been as a result of possibly a combination of rock and soil weathering as well as other domestic wastes and vehicle washout. Table 4b shows summary of factor analyses of the trace elements in Ala river sediments.

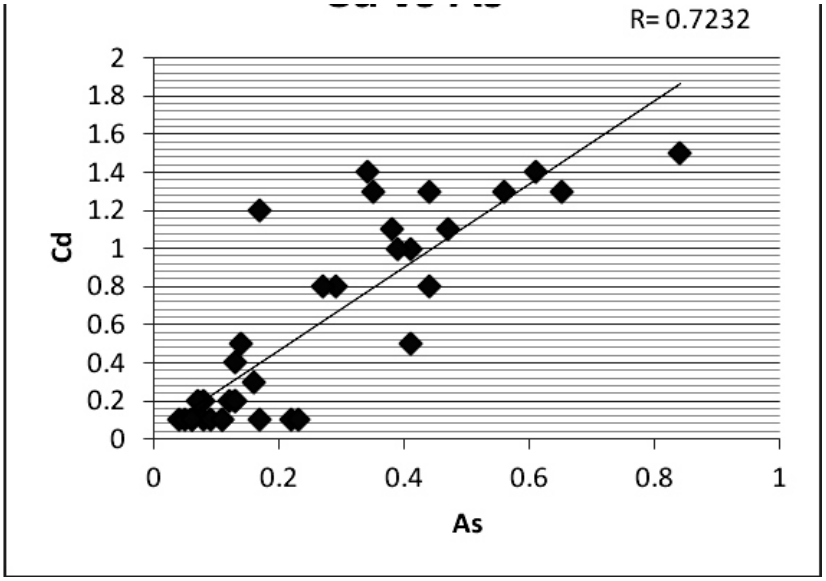
Further Application of the three component plot of principal component analyses (PCA) enabled us to find out further that the same trace elements clustered close to each other just like in the factor analyses i.e. those elements that are likely to be from analogous sources cluster together as seen in Figure 5. For the three component loading, Cu, Pb, Zn, As, Sr, Cd and Cr loaded close to each other suggesting similar source of enrichment. While Ni, V, Mn, and Co loaded close to each other also suggested the same source of enrichment. The third group of association were U, Th and La, suggesting entirely different source of enrichment in the Ala river sediment.



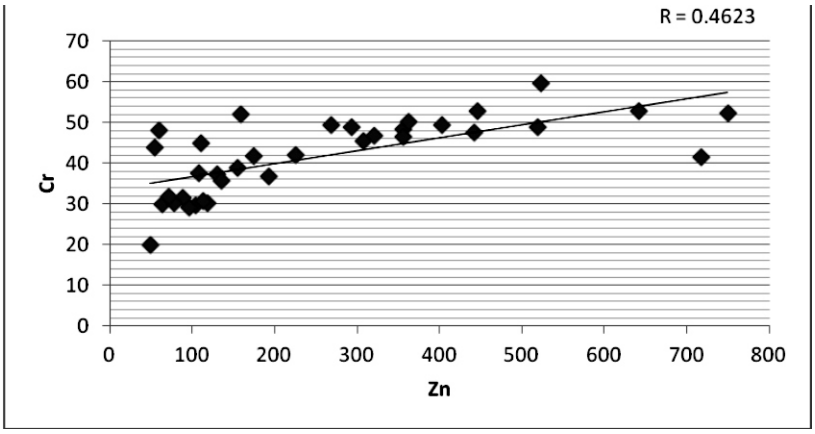
(a) Mn vs Co



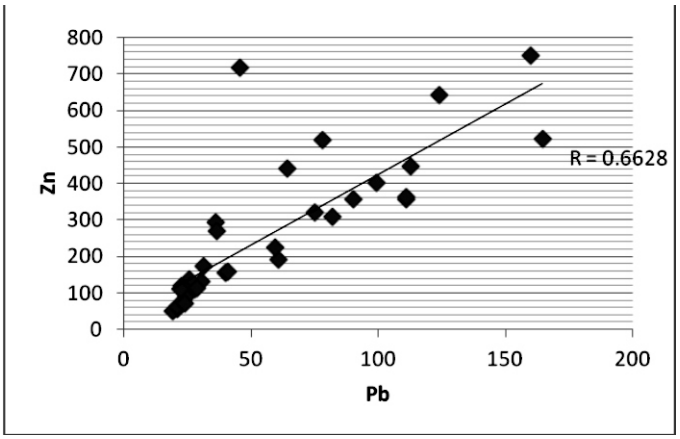
(b) Pb vs Cu



(c) Cd vs As



(d) Zn vs Cr



(e) Zn vs Pb

Fig. 4(a-e): Relationships Between Some of the Trace Elements in the Stream Sediments of the Study Area.

Table 3: Correlation Coefficients of Stream Sediments in the Study Area

	Cu	Pb	Zn	Ni	Co	Mn	As	U	Th	Sr	Cd	V	La	Cr
Cu	1													
Pb	**0.86**	1.00												
Zn	**0.88**	**0.81**	1.00											
Ni	0.17	-0.09	-0.12	1.00										
Co	-0.27	-0.50	-0.47	**0.81**	1.00									
Mn	-0.32	-0.37	-0.38	0.28	**0.64**	1.00								
As	**0.79**	**0.76**	**0.82**	0.08	-0.25	-0.17	1.00							
U	-0.07	0.08	-0.19	0.01	-0.07	-0.13	-0.20	1.00						
Th	-0.57	-0.38	-0.63	-0.10	-0.01	-0.08	-0.46	**0.49**	1.00					
Sr	**0.87**	**0.79**	**0.89**	0.12	-0.28	-0.28	**0.82**	-0.08	-0.53	1.00				
Cd	**0.93**	**0.92**	**0.95**	-0.10	-0.49	-0.37	**0.85**	-0.10	-0.57	**0.87**	1.00			
V	-0.04	-0.27	-0.30	**0.92**	**0.87**	0.36	-0.07	-0.02	-0.07	-0.09	-0.29	1.00		
La	-0.44	-0.34	-0.51	0.01	0.05	-0.10	-0.38	**0.59**	**0.86**	-0.46	-0.47	0.02	1.00	
Cr	**0.72**	**0.72**	0.68	0.31	-0.14	-0.29	0.74	-0.14	-0.42	**0.73**	**0.74**	0.13	-0.37	1

Table 4a: Principal Component Analyses of the Trace Elements in Ala River Sediments.

Element	F 1	F 2	F 3	F 4
Cu	0.92	0.16	0.19	0.05
Pb	0.89	-0.13	0.24	0.15
Zn	0.96	-0.05	-0.03	0.03
Ni	-0.06	0.88	0.41	-0.15
Co	-0.46	0.86	0.09	0.08
Mn	-0.38	0.53	-0.29	0.65
As	0.86	0.15	0.12	0.10
U	-0.20	-0.29	0.75	0.38
Th	-0.64	-0.40	0.52	-0.04
Sr	0.91	0.13	0.15	0.07
Cd	0.98	-0.07	0.07	0.09
V	-0.25	0.88	0.29	-0.15
La	-0.57	-0.33	0.66	0.01
Cr	0.78	0.28	0.27	-0.21
Eigen Value	6.9049	3.1026	1.8113	0.71055
% of Variance	49.321	22.162	12.9379	5.07534
Cumulative %	49.321	71.482	84.4203	89.4956

Table 4b: Summary of Elemental Associations of the Four-Factor Model and the Eigen Value.

Factor	Element Association	Eigen Value Ass %
R-1	Cu-Pb-Zn-As-Sr-Cd-Cr	49.32
R-2	Ni-Co-Mn-V	22.16
R-3	U-Th-La	12.94
R-4	Mn	5.08

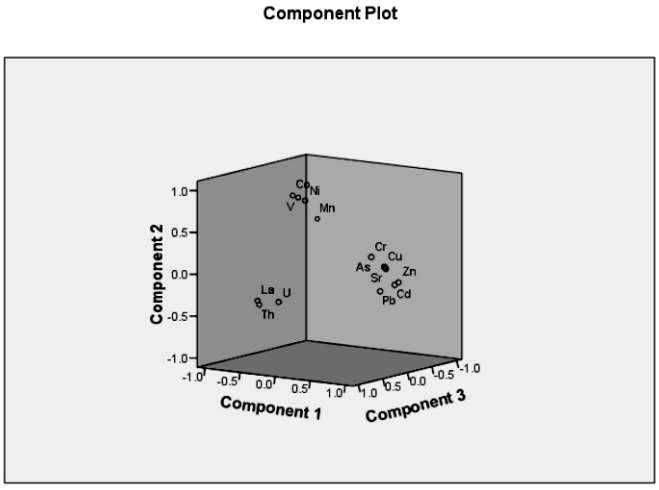


Fig. 5: Plot of the Three Principle Component for the Trace Elements in Ala River Sediments

The stream sediment quality assessment of the contamination status were evaluated using Geo-accumulation index. This method has been used successfully by researchers (Muller, 1979, Forstner and Wittmann, 1983; and Odewande and Abimbola, 2008) to determine the pollution status of soil. The formula commonly used for th evaluation of I-geo value is

$$\text{I-geo} = \text{Log}_2 Cn/1.5B_n$$

Where I-geo represents the geo-accumulation index, C_n represents the measured concentration of the element in the sediment, B_n represent the background value (where, for this paper, Bn is the average of both the granite rocks and the earth crust value) (Levinson,1974) and 1.5 is the background matrix correction. I-geo is classified into seven (7) classes (Muller, 1979), ranging from unpolluted to very highly polluted. The summary of the calculated I-geo values from selected metals in the study area is presented in Figure 5. The results of the I-geo revealed that Ala river sediment vary from unpolluted, for example in sample AKR/SS/12 for Cu and Pb to highly polluted, as observed in AKR/SS/2 for Zn. All the sediments that were observed to be moderately to highly polluted are those samples obtain from locations close to where high human activities were taking place, like the commercial areas. This is in agreement with other published work with similar studies (Odewande and Abimbola, 2008; Abimbola et al. 2006; Olatunji and Abimbola, 2010).

Table 5: Summary of Geo-accumulation Index Calculated for Selected Metals in Ala River Sediments.

Metal	I-geo Range	Interpretation
Cu	0.76 - 3.89	Unpolluted – highly polluted
Pb	0.66 – 3.72	Unpolluted – highly polluted
Zn	2.39 – 6.25	Moderately – very highly polluted

15

CONCLUSION

The Ala River sediments have been evaluated for pollution using a total of 36 sediment samples in which the concentrations of Cu, Pb, Zn, Cr, Ni, Mn, As, Th, La, Sr, Co, V, Th, U and Cd were determined. Although these elements could be found naturally in sediments, the relatively high concentrations of Mn, Zn and Pb at some locations suggested that their enrichment in the sediments could be traced to anthropogenic sources. The relatively high concentrations of Mn, Zn and Pb, indicated that these metals may be potentially hazardous to the aquatic environment. Elevated metal concentrations were found at locations close to active commercial centers and human waste discharges into the stream, indicating anthropogenic input. Comparing the results obtained in the sediments with the control result from the average earth crust and the various rocks especially for Mn, Zn and Pb, there was a considerable elemental enrichment in the stream sediments of between 4 to 6 folds in most of the locations. Further pollution level quantification of the trace metals revealed that Pb, Zn and Cu had reached significant pollutant status in the Ala River sediments as observed in the calculated geo-accumulation index in which some of the metals had status ranging from moderately contaminated to very highly contaminated.

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