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HEAVY METAL POLLUTION AND ECOLOGICAL RISK ASSESSMENT IN THE SEDIMENTS OF RIVER KADUNA, NIGERIA

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ABSTRACT

This study evaluated the pollution level and ecological risk of Cd, Cu, Cr, Pb and, Zn in sediments of River Kaduna. The total metal concentrations were analyzed using Atomic Absorption Spectrophotometry (AAS), while the pollution level and ecological risk of the metals were evaluated using a combination of geo-accumulation index (I_{geo}), contamination factor (CF), pollution load index (PLI), Nemerow pollution index (NPI), and potential ecological risk index. While Cd was below detection limits, the level of other heavy metals in all the sites followed the order: Zn > Cr > Cu > Pb. Heavy metals in the sediment can be classified as unpolluted to moderately polluted ($I_{geo} < 2$), with the pollution level following the order of Cr > Cu > Pb > Zn. While PLI classified the sites as polluted, NPI further subdivided the pollution level of the sites as precaution domain, slightly polluted domain, and moderately polluted domain. Pollution of the sites decreased from March to September at sites MU and JI but increased at NK. The same pattern was also observed for the ecological risk of the sites. However, none of the metals posed an ecological risk in the area as the risk factors were all below the lower threshold ($E^r < 40$), and there was also a low risk to the local ecosystem at all the sites from the studied metals (RI < 110). The metals showed seasonal and spatial variation with levels that did not pose a serious threat in the area.

Keywords: Heavy metals, ecological risk, pollution, sediment, river Kaduna, index

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INTRODUCTION

Environmental pollution by heavy metals is of great concern around the world and has been listed as priority pollutants to control (Tóth *et al*., 2016). Many of these heavy metals are necessary for the normal function of plants and animals, while some (like As, Cd, Pb, and Hg) have no known function in plants and animals (Mertz, 1981). However, they are generally non-biodegradable and even those that are considered as essential elements can accumulate to a toxic level (Nowrouzi and Pourkhabbaz, 2014).

Sources of heavy metals in the environment can be both natural/lithogenic sources (e.g. weathering of rocks, soil erosion, volcanic activities, etc.) and anthropogenic sources (e.g. agricultural activities, industrial activities, mining, vehicular emission, etc.). Heavy metals from both sources contaminate soil, water and, sediments (Ali *et al*., 2019). Sediments are important sinks for pollutants like organic pollutants and heavy metals (Yin *et al*., 2011). More than 90 % of the total heavy metals load in the aquatic environments is bound to suspended particulate matter and sediments (Islam *et al*., 2017). Heavy metals bound to suspended particles subsequently accumulate in sediments through the processes of precipitation, coprecipitation, chelation, and biological effects, hence the highest concentration of toxic metals in the aquatic environment is found in sediments (Hamuna and Wanimbo, 2021). As a sink for pollutants, sediments have been used as a sensitive indicator for monitoring the contamination level or quality of an aquatic system (Singovszka and Balintova, 2016; Mandeng *et al.,* 2019; El-Amier *et al*., 2021).

The study of heavy metals in sediments is vital because of their significant impact on human life and the aquatic ecosystem. Sediment is both a habitat for aquatic organisms as well as a source and a sink of contaminants (Akpan and Thompson, 2013; El Madanil and Hacht, 2017). It provides nutrients and substrates for micro and macro flora and fauna and can pose an ecological risk to benthos (Ali *et al*., 2019; Bat and Ozkan, 2019). Heavy metals in sediments cause a decline in ecosystem productivity, loss of biological biodiversity, alteration of habitats, and contamination of aquatic biota (Yunus *et al*., 2020). Physicochemical parameters such as a change in pH, redox potential, and degradation of organic matter, significantly control the mobility and availability of heavy metals in aquatic environment. Due to changes in these parameters, the heavy metal-loaded sediment can cause secondary pollution of the underlying water by releasing back the heavy metals into the water bodies, and consequently to living organisms including man (Yunus *et al*., 2020; Liu *et al*., 2021; Astatkie *et al*., 2021).

Heavy metals have the ability of bioaccumulation and biomagnification in organisms across the trophic level, thereby posing a greater risk to organisms at the higher trophic levels in the food chain (Chindah *et al*., 2009; Ali *et al*., 2019). Severe adverse effects of heavy metals on humans have been documented (Yunus *et al*., 2020; Kuang *et al*., 2021). The results of the sediment contamination study provide information to policymakers and resource managers that can inform decision-making (Ntakirutimana *et al*., 2013). Different evaluation methods have been applied to evaluate heavy metal pollution in soil and sediments. Such methods include geochemical normalization methods (e.g. sediment quality guidelines, geo-accumulation index, pollution load index, contamination factor, enrichment factor, ecological risk index) (Lodhaya *et al*., 2017; Ngwoke *et al*., 2019; Algül and Beyhan, 2020; Astatkie *et al*., 2021; Onoyima, 2021); multivariate statistical methods (Okibe *et al*., 2020, Yisa *et al*., 2011); Monte Carlo simulation method (Chen *et al*., 2019; Kuang *et al*., 2021).

The present study aims to use geo-accumulation index, contamination factor, pollution load index, Nemerow pollution index, and ecological risk index to evaluate the contamination level and risk of some heavy metals in sediment of River Kaduna. While a single index has been applied for heavy metals pollution analysis, the use of several independent indices provides a comprehensive assessment and prevents bias that arises from using a single index (Wang *et al*., 2019).

MATERIALS AND METHODS

Sediment Sampling and Pre-treatment

Nine (9) composite samples were collected at the banks of each of the three selected sample sites and were immediately put in sterilized polythene bags. The samples were transported to the laboratory, air-dried, lump samples were gently crushed and sieved to a particle size of 2.00 mm and transferred into amber glass bottles sealed and labeled before storing in a refrigerator (SAEFL, 2003). This is to ensure the reproducibility of results and precision.

Sediment Digestion and Heavy metal Analysis

The digestion of the sediment sample was done by dissolving 1.00 g of the dried powdered sediment sample in a clean 100 cm^3 beaker followed by the addition of 20 cm^3 concentrated HCl, 5.00 cm^3 concentrated HNO_3 and 2.00 cm^3 concentrated HF. The mixture was then heated to boil for one hour, filtered hot, and made up to the mark with distilled water in a 100.00 cm^3 volumetric flask and finally, the heavy metals were analyzed using AAS (US EPA, 1999).

Geo-accumulation index

The Geo-accumulation Index (Igeo), was introduced by Muller, (1969) for determining the extent of metal accumulation in soil and sediments, and has been used by various workers for their studies. Igeo is mathematically expressed as:

$$Igeo = \log_2 \frac{C_n}{1.5C_B} \text{ .... [1]}$$

Where C_n is the concentration of the metal in the sediment, C_B is the geochemical background value. Factor 1.5 is incorporated in the relationship to account for possible variation in background data due to the lithogenic effect. The geo-accumulation index (Igeo) scale consists of seven grades (0 –6) ranging from unpolluted to highly polluted (Table 1).

Contamination Factor (CF) and Pollution Load Index (PLI)

PLI evaluates pollution for a particular site following the method proposed by Tomlinson *et al*., (1980), which can be stated as follows:

$$PLI = \sqrt[n]{CF_1\ x\ CF_2\ x\ CF_3\ x \ldots CF_n} \text{ .... [2]}$$

Where n is the number of metals and CF is the contamination factor, which compares the measured concentration (C_n) with the background value (C_B)

$$CF = \frac{C_n}{C_B} \text{ .... [(3)]}$$

Nemerow Pollution Index (NPI)

NPI is an integrated pollution index that highlight the importance of the most contaminated element, and has been widely used to assess the quality of soil and sediment (Jiang *et al*., 2014; Ikpe *et al*., 2018)

$$NPI = \frac{\sqrt{\left(\frac{\sum_{i=1}^{n} CF}{n}\right)^2 + (CF_{max})^2}}{2} \text{ .... [4]}$$

Where: NPI = Nemerow pollution index
CF = contamination factor
n = number of elements analysed
CF_{max} = the maximum value of the contamination factor of the investigated heavy metals
The quality of sediment is classified into five categories as shown in Table 1.

Ecological risk index

The Potential Ecological Risk Index (RI) was originally introduced by Hakanson, (1980) to assess the degree of heavy metal pollution in soil, according to the toxicity of metals and the response of the environment. RI could evaluate ecological risk caused by toxic metals comprehensively. The calculating methods of RI are listed below:

$$F_i = \frac{C_n}{C_r} \text{ .... [5]}$$

$$E^i_r = T^i_r \text{ x } F_i, \text{ .... [6]}$$

$$RI = \sum\nolimits^{n}_{i=1} E^i_r \text{ ..... [7]}$$

Where F_i is the single metal pollution index; C_n is the concentration of metal in the samples; C_r is the reference value for the metal; E^i_r is the monomial potential ecological risk factor; T^i_r is the metal toxic response factor according to Hakanson, (1980). The values for each element are in the order Zn = 1 < Cr = 2 < Cu = Pb = 5 < Cd = 30. RI is the potential ecological risk caused by the overall contamination. There are four categories of RI and five categories of E^i_r as shown in Table 1.

Table 1: Different assessment indices and their grades

Geo-Accumulation Index (Igeo)

Igeo value	pollution status
Igeo<0	unpolluted
0<Igeo≤1	unpolluted to moderately polluted
1 < Igeo ≤ 2	moderately polluted
2 < Igeo ≤ 3	moderately to heavily polluted
3 < Igeo ≤ 4	heavily polluted
4 < Igeo ≤ 5	heavily to extremely polluted
Igeo > 5	extremely polluted

Contamination Factor (CF)

Range	Status
CF < 1	low Contamination
1 ≤ CF < 3	Moderate Contamination
3 ≤CF < 6	Considerable contamination
CF ≥ 6	very high contamination

Pollution Load Index (PLI)

Range	Status
PLI > 1	Polluted
PLI < 1	Not polluted
PLI = 1	close to background level

Nemerow Pollution Index (NPI)

Range	Status
NPI < 0.7	safe domain
0.7≤NPI <1	precaution domain
1 ≤ NPI < 2	slightly polluted domain
2 ≤ NPI < 3	Moderately Polluted
NPI > 3	seriously polluted

Monomial Potential Ecological Risk (E^i_r)

Range	Status
$E^i_r < 40$	low risk
$40 \leq E^i_r < 80$	moderate risk
$80 \leq E^i_r < 180$	considerable risk
$160 \leq E^i_r < 320$	high risk
$E^i_r \geq 320$	very high risk

Total Potential Ecological risk (RI)

Range	Status
RI < 110	low risk
110≤RI<200	moderate risk
200 ≤RI<400	Considerable risk
RI ≥ 400	very high risk

RESULTS

The mean concentrations of the heavy metals in the sediment of River Kaduna are presented in Figure 1. The results show that while Cd was below the detection limit in all the samples, the level of other heavy metals in all the sites follows the decreasing order: Zn > Cr > Cu > Pb. Zn has the highest value of 52.56±4.06 mg/kg at JI (March), and the lowest mean value of 20.67±0.73 mg/kg at NK (March).

Cr was the second most abundant among the studied heavy metals in the area. The mean concentration peaked at JI (March) (47.22±2.27 mg/kg), with the lowest value of 21.22±0.40 mg/kg at NK (March). Cu has the highest mean concentration of 23.44±1.46 mg/kg at JI (March) and the lowest mean value of 9.78±0.49 mg/kg at NK (March).

The pollution level of heavy metals in this study was assessed with a combination of geo-accumulation index (I_{geo}), contamination factor (CF), pollution load index (PLI), and Nemerow pollution index (NPI). CF and I_{geo} assess the contamination degree of individual heavy metals, while PLI and NPI evaluate the combined pollution of multiple heavy metals in sediments. The results of the I_{geo} was presented in Figure 2. The ecological risk assessment results for heavy metals in the sediment were summarized in Figure. 3.

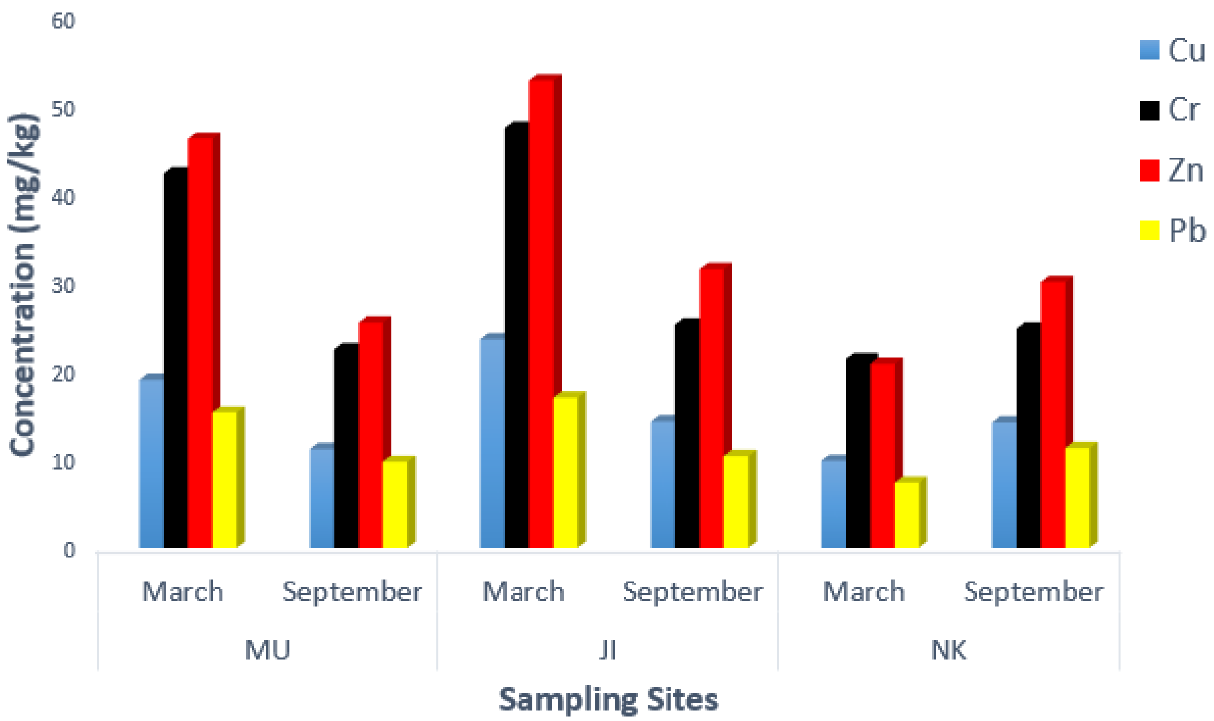

Figure 1: Mean concentrations (mg/kg) of heavy metals at the sites in March and September

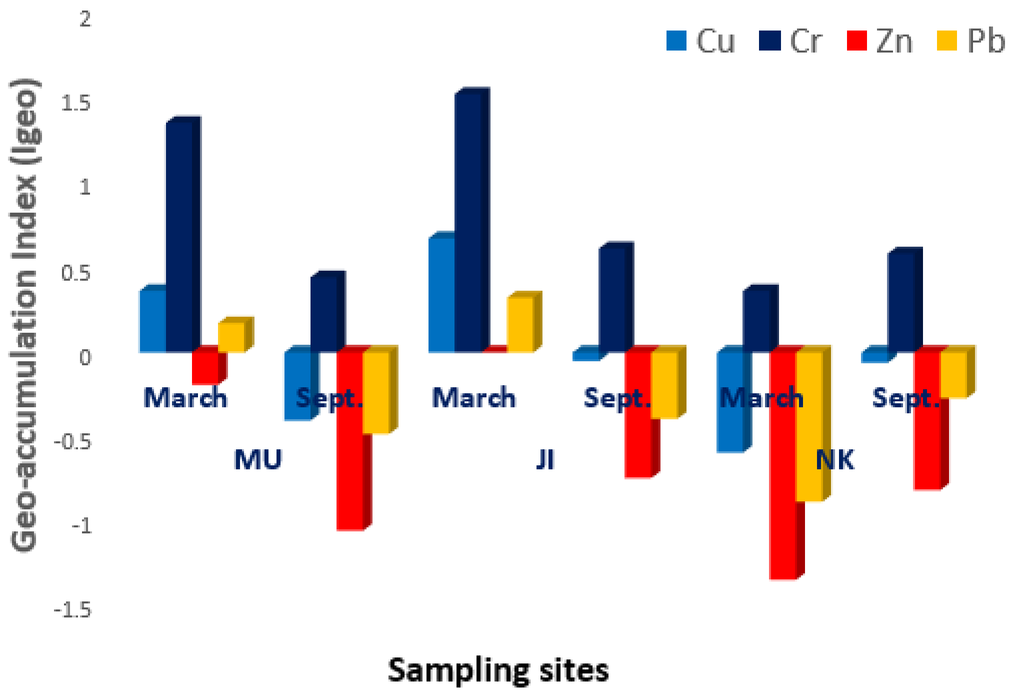

Figure 2: Geo-accumulation index of heavy metals in the sediment of the study area

Table 2: Results of Contamination Factor (CF), Pollution Load Index (PLI), and Nemerow Pollution Index (NPI)

	MU		JI		NK	
	March	Sept.	March	Sept.	March	Sept.
CF (Cu)	1.92	1.13	2.39	1.45	1.00	1.44
CF (Cr)	3.83	2.03	4.29	2.28	1.93	2.24
CF (Zn)	1.31	0.72	1.50	0.90	0.59	0.85
CF (Pb)	1.69	1.07	1.88	1.15	0.81	1.25
PLI	2.01	1.15	2.32	1.36	0.98	1.35
NPI	2.21	1.19	2.49	1.35	1.11	1.33

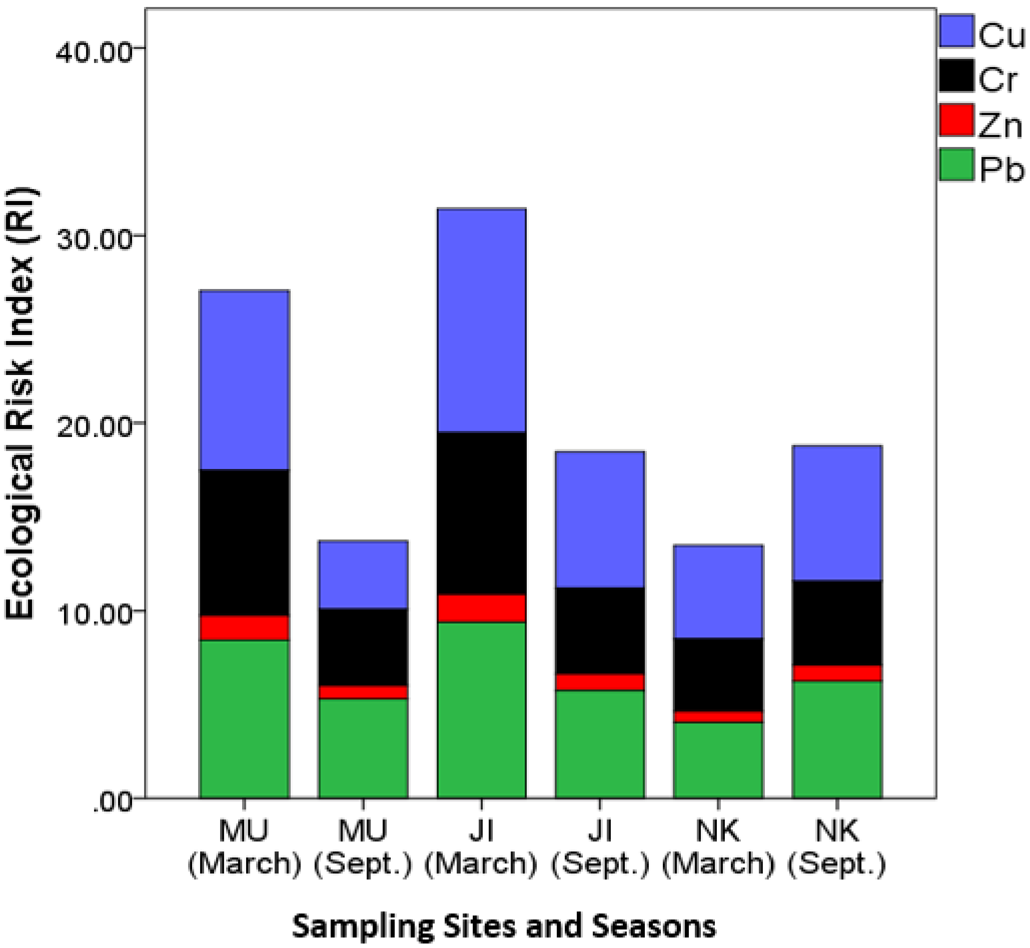

Figure 3: Contributions of each metal to total ecological risk

DISCUSSION

Mean concentrations of heavy metals in sediment

Many types of research involving zinc showed that its level in sediments was higher than other metals (Yisa *et al*., 2011; Hamuna and Wanimbo, 2021; Kuang *et al*., 2021). Both natural/geological and anthropogenic sources contribute to the high concentration of Zn in sediments. It is the 24th most abundant element on earth, with about 0.007 % of the earth's crust (Krebs, 2006), and can naturally enrich the sediments from several minerals such as ZnS, $ZnCO_3$, ZnO, etc. (Al-Edresy *et al*., 2019).

Anthropogenic input of Zn includes metal works, battery and printing materials, agricultural activities, etc. (Al-Edresy *et al.*, 2019; Yunus *et al.*, 2020). Although Zn has low toxicity, it is present in high concentrations and an easily mobilizable form in sediment (Tamás and Farsang, 2016). The values of Zn obtained in the present study are lower than that reported by Liu et al., (2021) and El-Amier *et al.*, (2021), but higher than that of Kassegne *et al.*, (2018) and Aigberua *et al.*, (2020).

Sources of Cr include weathering of minerals, atmospheric deposition from coal-burning dust, electroplating, and laboratory effluents (Cui *et al.*, 2019; Astatkie *et al.*, 2021). Cr (VI) has low mobility under moderately oxidizing and reducing conditions and nearly neutral pH (Decena *et al.*, 2018). The values of Cr obtained in this study are lower than that obtained by Cui *et al.*, (2019), comparable to the results of Liu *et al.*, (2021), but higher than that of Hamuna and Wanimbo, (2021).

Cu has the highest mean concentration of 23.44±1.46 mg/kg at JI (March) and the lowest mean value of 9.78±0.49 mg/kg at NK (March). These values are comparable to those reported by Kuang *et al.*, (2021) and Akpan and Thompson, (2013) in Asogha beach, Cross River, Nigeria. In addition to naturals/geological sources, Cu can anthropogenically pollute the environment through electrical equipment, chemicals, paints, agricultural pesticides and preservatives, vehicular emission and brake pad wear, etc. (Cui *et al.*, (2019; Al-Edresy *et al*, 2019). Pb recorded the lowest level in the sediment of River Kaduna, with a maximum value of 16.89±0.48 kg/mg at JI (March) and a minimum value of 7.33±0.17 kg/mg at NK. Low values of Pb in sediment were also reported in Niger Delta, Nigeria by Aigberua *et al.*, (2020) (mean: 7.50 kg/mg), in Bangladesh by Jewel *et al*, (2020) (6.43 mg/kg), and in Uganda by Sekabira *et al.*, (2010) (10.00±0.98 mg/kg). Anthropogenic input of Pb outweighs the natural input and includes municipal waste, gasoline stations and vehicular emissions, construction industries, etc. (Al-Edresy *et al*, 2019; Astatkie *et al.*, 2021).

There were significant differences in the level of heavy metals among seasons and sampling sites in the sediment. The spatial variation can be due to varying topography, hydrology, geology, and land use (Islam *et al.*, 2017; Zarezadeh *et al.*, 2017). The result shows that all the metals decreased in level from March to September at both Mu and JI sites, but increased from March to September at site NK. Seasonal variation in the level of heavy metals in sediment is affected by physical processes, biogeochemistry, geomorphological setup, and change in water volume (Kumer *et al.*, 2015; Mortazavi and Hatami, 2018). Higher concentrations of heavy metals at Mu and JI in March can be attributed to lower water levels during this season. Lower water levels can lead to the precipitation of contaminants in sediments (Silva *et al.*, 2019; Kormoker *et al.*, 2019). The seasonal temperature change can also lead to shifting in organic matter decomposition, varying pH, and consequently varying heavy metal levels (Bazzi, 2014). A similar observation was made by Jewel *et al.*, (2020) and Islam *et al.*, (2015). However, the increase in metal level from March to September at site NK indicates that water level is not the only factor controlling the metal level in sediment. Metal concentration in sediment is influenced by adjacent land use type (Ikpe *et al.*, 2018; Cui *et al.*, 2019). The effect of the point source can also alter the seasonal trend of heavy metals in sediments (Kormoker *et al.*, 2019). Okibe *et al.*, (2020) applied multivariate to assess the pollution status of River Kaduna. The result confirms the significant difference between the two sampling seasons.

Pollution levels of heavy metals in the sediment

Based on the results (Figure 2), the studied heavy metals in the sediment can be classified as unpolluted to moderately polluted ($I_{geio} < 2$) (Table 1). Cu falls under the unpolluted class at Mu (Sept.), JI (Sept.), and NK (March and Sept.), but was under unpolluted/moderately polluted class at MU (March) and JI (March). Cr moderately polluted the sediment at MU (March) and JI (March) but was under unpolluted/moderately polluted class at other sites/seasons. While Zn was in the unpolluted class in all the results, Pb was also in the unpolluted class except for MU (March) and JI (March), where it was in the unpolluted/moderately polluted class. Based on the geo-accumulation index, the metal pollution can be arranged in the following decreasing order: Cr > Cu > Pb > Zn. This order did not

follow the concentration level because I_{geo} considers the background values of the metals.
Contamination factor (CF) is a less conservative way of assessing the contamination level of heavy metals. The results of the contamination factor in this study (Table 2) placed the heavy metal in the sediment under three categories: low contamination, moderate contamination, and considerable contamination. Only Cr was in the considerable contamination class at Mu (March) and JI (March). The low contamination class includes Zn at MU (Sept.), JI (Sept.), NK (March and Sept.), and Pb at NK (March), while the remaining results were in the moderately contaminated class.
The results of the PLI placed all the sites as polluted at the two studied seasons except NK in March, which showed the background values of the metals. NPI further subdivides the pollution level of the sites as follows: NK was in the precaution domain class in March, all the sites were in the slightly polluted domain in September, while MU and JI were in the moderately polluted domain in March. The order of pollution of the sites in March was JI > MU > NK, while in September it was NK = JI > MU. The results also showed that the pollution of the sites decreased from March to September at MU and JI, but increased at NK.

Ecological risk assessment

The monomial potential ecological risk index (or potential ecological risk factor) (E_r), introduces a toxic response factor for a given pollutant (Kormoker *et al*., 2019). It was found that the average monomial risk factors (E_r) were ranked in the following order: Cu > Pb > Cr > Zn. All the metals constituted low ecological risk at the studied sites within the study period as the risk factors were all below the lower threshold (E_r < 40). The average contribution of each metal to total ecological risk is as follows: Cu (37.34 %), Pb (31.35 %), Cr (26. 61 %), and Zn (4.70 %). Although Zn had the highest concentration in the area, it has the least ecological risk factor. This is because Zn has a low toxic response factor and risk is a combination of pollution and toxic response factor. Cr, on the other hand, has the highest pollution level but constituted a lower risk than Cu due to lower toxic response factors.
RI was used to evaluate the total risk caused by all the studied metals. RI represents the sensitivity of the biological community to the toxic metal and illustrates the potential ecological risk caused by the overall metals (Islam *et al*., 2017). The result shows there was a low risk to the local ecosystem at all the sites from the studied metals (RI < 110). A high ecological risk index means that marine or benthic organisms are exposed to environmental risk. The order of ecological risk of the sites in March is JI > MU > NK, while in September, it was NK > JI > MU.

CONCLUSION

The use of integrated indices in this study gave a clear status of heavy metals concentration, pollution, and risk in the sediment of River Kaduna. The concentrations of the metals were in the order of Zn > Cr > Cu > Pb, while introducing background values showed the pollution level was in the order of Cr > Cu > Pb > Zn, and when the toxic response factor of each metal was incorporated the risk was found to be in the order of Cu > Pb > Cr > Zn. Due to factors such as a change in water level, land use type, change in temperature, geomorphological setup, and other physicochemical processes, the metals showed seasonal and spatial variations with levels that did not pose a serious threat in the area.

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