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POLLUTION STATUS AND ECOLOGICAL RISK OF HEAVY METALS IN SURFACE WATER AND SEDIMENT OF THE COASTAL CREEKS OF ONDO STATE, NIGERIA

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Abstract

Despite the irreplaceable value of the aquatic ecosystem to livelihoods, the continuous release of heavy metals into the marine environment have remained an issue of global concern due to their bioaccumulation, and potential toxicity in biological systems and humans. This study investigated the Pollution Status and Ecological Risk of Heavy Metals in Surface Water and Sediment of the Coastal Creeks of Ondo State. Water and sediment samples were collected from four stations between June, 2019 and April, 2021. Heavy metals in the samples were analyzed with GBC Savant AA Sigma Flame Atomic Absorption Spectrometer (AAS). The results of the mean level of heavy metals were in the order; Fe>Zn>Cu> Ni> Co>Pb> Cd>Cr for surface water; and Fe> Cu> Co> Ni >Zn > Pb > Cr> Cd for sediment. The single-factor pollution index revealed slight to serious pollution of the water, and low to moderate pollution of the sediment. The degree of contamination (Cd) and the geoaccumulation factors (Igeo) of individual metals were also low. The ecological risk (Er) of individual metal showed low risk in the sediment, except for Cadmium that showed moderate risk (40<Eir ≤ 80) across the stations. On the other hand, the potential ecological risk (RI) revealed low risk (RI <150) of heavy metals in the sediment. The enrichment factors (Ef) for individual metals were greater than 40 (Ef \geq 40), which suggests extremely high enrichment of heavy metal in the sediment through anthropogenic sources. This study provided insights into the pollution status of the surface water and sediment of the coastal creeks of Ondo state and it associate potential ecological risks.

Keywords: Coastal waters; sediments; heavy metals, Pollution status; ecological risk.

Introduction.

Continuous increase in urbanization and industrialization are the major reasons for the continuous discharge of large quantities of heavy metals from anthropogenic sources into the marine ecosystem, has resulted to surface water pollution and devastating threat to aquatic biota and human health (Garnero *et al.*, 2010; Ajani *et al*., 2021). Heavy metals enter the sediments in marine environment via numerous pathways, including fertilization, irrigation, runoff, rivers, deposition of the atmosphere, and land sources, where metal production occurred

from by-products of crude oil refining and metal mining (Ajayi *et al*., 20211).

Heavy metals in water are enriched in sediments and organisms through various ways, which will cause unavoidable harm to the organism (Ediagbonya and Adesokan, 2019; Ediagbonya *et al*., 2022). In the assessment system of water environmental pollution, heavy metals have already become the important factor of the marine environmental quality evaluation (Ray *et al*., 2013; Mustapha *et al*., 2021; Loto *et al*., 2022). Some metals, such as Fe, Cr, Cu, Ni, Mn, and Zn are necessary for marine organisms, but they may be toxic if present in high concentrations. On the other hand, some metals are considered mostly toxic to the marine life, even if they exist in a little quantity (Jumbo *et al*., 2015), for example Hg, Pb, Cd, and Ag. Sediments are regarded as the main store and final receiver for the metals discharged into the ecosystem (Mustapha *et al*., 2021).

Heavy metals are known to be one of the major inorganic pollution sources, with a significant negative impact on marine ecosystems (Bodin *et al*., 2013). They show increasing concentrations in sediments through complex physical and chemical adsorption mechanisms depending on the nature of the sediment matrix and the properties of the adsorbed compounds there-in (Ibanga *et al*., 2019). They are considered a potential secondary water pollution source with changes in environmental conditions such as pH, redox potential, organic matter content and temperature (Fahad *et al*., 2015). The content of heavy metals in sediments is usually three to six orders of magnitude higher than that in water, which implies that sediment is the main storage reservoir of heavy metals in water environment and plays an important indicator role for water pollution (Ajani *et al*., 2021). Currently,

various indexes have been developed to assess environmental risks for heavy metals in sediments based on their total contents, bioavailability, and toxicity (Liao *et al*., 2012; Bubu *et al*., 2018).

Heavy metal risk assessment methods based on total concentration calculation have been widely proposed (Xia *et al*., 2020), mainly including two categories: one is related to background values, the other is related to sediment quality guidelines. Background values mainly include three types: soil background value in each investigated area (BV), control value of surface sediment (Cvs, samples collected from the study area that are uncontaminated), and control value of deep sediment (Cvd, samples collected from the bottom of sedimentary column). indices help to determine whether the accumulation of heavy metals was due to natural processes or was the result of anthropogenic activities (Sutherland, 2000; Elias and Gbadegesin 2011; Caeiro *et al*. 2015). Further, pollution indices have a great importance for monitoring soil quality and ensuring future sustainability, especially in the case of agroecosystems (Ogunkunle and Fatoba 2013; Ripin *et al*. 2014).

The increasing needs to meet the food supply of growing population has put a lot of pressure on coastal and marine resources, therefore, the utilization of wastewater for the production of aquatic biota has created great harm to human through the food chain contamination (Iqbal *et al*., 2020).

Mahin Lagoon is one of the Lagos lagoon complex systems in the Gulf of Guinea coast, West Africa (Loto *et al*., 2023). It meets a lot of social economic needs of the people such as fishing, sand mining, transportation, etc. There are reported proliferations of coastal settlements that have led to the generation of industrial and domestic anthropogenic effluents which eventually find their way into

the Mahin lagoon as an ultimate sink (Loto et *al.,* 2023). This study aimed to investigate the level of concentrations, the pollution status, and the potential ecological risk of heavy metals in surface water and sediment of the coastal creeks in Ondo State, Nigeria.

Materials and Methods

Study Area

The coastal area of Ondo State lies on Latitude 5° 50'N – 6° 09'N and Longitude 4 $^{\circ}$ $45'E - 5°05'E$ (Ajibare, 2014). The area is occupied by the Ilajes in linear settlements along the coast with a fast-growing population, most of whom utilize the water for various purposes (Agunbiade *et al.* 2010). The study area witnesses high economic activities with active oil exploration, fishing and other agricultural activities, trading and boat making. Boat transportation appears to be the only means of transportation of people and goods in the area. These activities, alongside other municipal use tend to pollute the water and its underlying sediments Mangrove swamp is the dominant vegetation type in this area, especially the red mangrove Rhizophora racemose and the white mangrove Avicennia spp, typical of swamps. A striking feature of vegetation in the area is the desiccation induced by marine water incursion into about 10,000 hectares of freshwater swamp forest (Ajibare *et al*., 2018). The terrain is characterized by near sea level swamp flat at the estuary which gently rises northward. The area is drained by many perennial streams and rivers, that traverse several settlements of the coast, and empties into the open ocean through the estuary with exchanges of water between the ocean and the coastline (Loto *et al.* 2022). There is also the prevalence of erosion gullies along the river banks, shoreline and coastlines. Mud crack is a common sedimentary structure found in

the area during the dry season, usually formed by dried saturated mud.

Four (4) major fishing communities, Ugbo, Ugbonla, Ayetoro and Oroto were selected in Ilaje Local Government Area of Ondo State. The site selection was based on the population/aggregation of fishing families/possible anthropogenic inputs, geographic distribution, catch volume and species diversities of the fish catches in the area.

Samples' collection and Analysis Water samples

A total of Two Hundred and Eight-Eight (288) samples of water were collected with 1L sampling bottles in the entire sampling periods (24 months). Samples were collected bimonthly at each station and transported in ice chest to the Laboratory for the determination of heavy metals (Zn, Pb, Fe, Cu, Cr, Cd, Ni and Co) analysis. 25 ml of each water sample was digested with 15 ml of HNO₃ in a fume cupboard at 130°C until 2–5 mL remained in the beaker. The water digests were then filtered through Whatman no. 41 filter paper and then made to 50 mL volume using deionized water. Sampling and analysis were conducted according to APHA (APHA, 2000).

Sediment samples:

A total of Two Hundred and Eight-Eight (288) samples of sediment were collected in the entire sampling periods (24 months). Samples were collected bimonthly at each station, using the Van Veen grab (0.5 cm^2) , and kept in foil. The samples were placed in ice-chest and transported to the laboratory for analyses. Samples were air dried and after homogenization using pestle and mortar, they are passed through a 2 mm mesh screen and stored in polyethylene bags. About 2.0 g portion of dried sediment were digested in 15 cm3 of tri- acid mixture $(HNO₃, HCl and)$ $H₂SO₄$, as 5:1:1 ratio) at 80°C until the transparent solution appeared (Thomas and Mohaideen, 2015). After cooling, the digested sample was filtered using Whatman No. 41 filter paper and the filtrate was finally maintained at 50 cm3 distilled water. The clear solution was then poured into sample bottles for reading in the Atomic Absorption Spectrometer. All the matrixes were analyzed for Zn, Pb, Fe, Cu, Cr, Cd, Ni and Co by ACCUSYS 211 Atomic Absorption spectrophotometer, using the respective lamps and wave lengths.

*T***he Single-Factor Pollution Index (Pi) Pollution Indices of Water**

The Single-Factor Pollution Index (Pi) was calculated as:

$$
P_i = \frac{C_i}{S_i} (Yan \text{ et al.}, 2015)
$$
 (1)

Where: P_i is the pollution index of pollution indicator in water,

 C_i is the concentration of the pollution indicator in water (mg/l)

 S_i is the permissible limit for the pollution indicator in water.

The single-factor pollution index (P_n) is classified into five grades, according to Yan *et al.,* (2015) (Table 2).

Figure 1: Map showing sampling locations in Coastal Creeks of Ondo state

Table 1: Standards for single-factor pollution index (*Pi***)**

Pi	Pollution grades				
< 0.4	Non-pollution				
$0.4 - 1.0$	Slight pollution				
$1.0 - 2.0$	Medium polluted				
$2.0 - 5.0$	Heavy polluted				
>5.0	Serious polluted				

$$
CPI = \frac{1}{n} \sum_{i=1}^{n} \frac{c_i}{s_i} \text{ (Tao } et \, al., 2011) \quad (2)
$$

Where: CPI = the comprehensive pollution index,

 C_i = concentration of the pollution indicator *i* (mg/l) ,

 S_i = permissible limit for the pollution indicator *i* in water

n = the number of analyzed pollution indicators.

CPI is classified according to Tao *et al.* (2011) into five water quality levels (Table 3).

Pollution and Ecological Risk Assessment of Heavy Metals in Sediment Single Pollution Index (PI)

An index that can be used to determine which heavy metal represents the highest threat for a soil environment is the Single Pollution Index (PI). *Cn*

$$
Pi = \frac{Cn}{GB} \tag{3}
$$

where Cn is the content of heavy metal in sediment and GB is values of the geochemical background.

The Nemerow Pollution Index $(PI_{Nemernw})$ was calculated as describes by Gong *et al.*, (2008), using equation (4). **Nemerow Pollution Index (PI** $_{\text{Nemero}}$)

$$
PI_{Nemerow} = \frac{\sqrt{\left(\frac{1}{n}\Sigma_{i-1}^n P I\right)^2 + P I^2 max}}{n}
$$
 (4)

$$
^{(4)}
$$

where PI—calculated values for the Single Pollution Index, PI max—maximum value for the Single Pollution Index of all heavy metals and n—the number of heavy metals. Based on $PI_{Nemerow}$, five classes of sediment quality were created (Table 3).

Contamination Factor (Cf)

Contamination factor Cf was calculated by dividing the concentration of metal in the sample by the concentration of the same metal in the reference or background area, as described by Hakanson (1980), using equation (5):

$$
Cf = \frac{Cs}{Cb} \tag{5}
$$

where Cf is the contamination factor, Cs is the metal concentration in the sample, and Cb is its background (reference) concentration.

Degree of Contamination (Cd)

The Degree of Contamination (Cd) of heavy metal in sediment was estimated according to Hakanson (1980), using equation (6):

$$
Cd = \sum_{i=1}^{n} Cf
$$
 (6)

where Cf—contamination factor and n—the number of analyzed heavy metals.

Geoaccumulation Index (Igeo)

The Geoaccumulation Index (Igeo) of heavy metals in the sediment was calculated according to Muller, (1969), using equation (7):

$$
Igeo = log_2 \left[\frac{cn}{1.5GB} \right] \tag{7}
$$

where Cn—concentration of individual heavy metal, GB—value of geochemical background and 1.5 is constant, allowing for an analysis of the variability of heavy metals as a result of natural processes. Igeo values are helpful to

divide sediment into quality classes (Muller, 1969).

Potential Ecological Risk (PER)

Potential ecological risk (PER) of heavy metals in the sediment was estimated according to Hakanson (1980), using equation (8):

$$
PER = \sum_{i=1}^{n} Er \tag{8}
$$

where n—the number of heavy metals and Er—single index of the ecological risk factor calculated based on the equation:

$$
E_r^i = T_r^i \times PI
$$
 (9)

where T_r^i is the toxicity response coefficient of an individual metal (Hakanson1980) and PI— calculated values for the Single Pollution Index. Based on the potential ecological risk, five classes of soil quality were distinguished (Table 3).

Enrichment Factor (EF)

The Enrichment Facto (EF) was calculated

as described by Sutherland (2000), using equation (10):

$$
EF = \frac{\frac{Cn}{LV} \, sample}{\frac{GE}{LV} \, background} \tag{10}
$$

where $[Ch / LV]$ sample–content of analyzed heavy metal (Cn) and one of the following metals Fe/Al/Ca/Ti/Sc/ Mn (LV) in the sample and $[Ch / LV]$ $_{background}$ -reference content of the analyzed heavy metal (Cn) and one of the following metals Fe/Al/Ca/Ti/Sc/Mn (LV). If the value of EF ranges from 0.5 to 1.5 (Table 3), it can be stated that the content of that particular heavy metal in the sediment is caused by natural processes. However, if the value of EF exceeds 1.5, there is a possibility that the heavy metal contamination occurred as a result of anthropogenic activities (Elias and Gbadegesin 2011).

Table 3: Pollution categories for each evaluation index

Source: Mobarak *et. al.* (2021)

Results

Heavy metals concentrations in surface water and sediment of the Coastal Creeks

The mean concentration of heavy metal in surface water as presented in Table 4, was in the order; Fe>Zn>Cu> Ni> Co>Pb> Cd>Cr. They varied significantly across the stations with analysis of variance (ANOVA). Further test using Duncan multiple tests showed significant difference (p<0.05) in Cr, Cd, Ni and Co, across the stations. However, Zn, Pb, Fe and Cu were not significantly different (p>0.05) across stations.

of Ondo State The mean concentration of heavy metal in sediment as presented in Table 5, were in in the order; Fe> Cu> Co> Ni >Zn > Pb > Cr> Cd. They varied significantly across the stations with analysis of variance (ANOVA). Further test using Duncan multiple tests showed significant difference $(p<0.05)$ in all the metals except copper, across station.

Mean value with same superscripts along the row were not significantly different ($p>0.05$).

Mean value with same superscripts along the row were not significantly different ($p>0.05$).

Pollution Status of Heavy metals in the surface water and sediment of the coastal creeks of Ondo State

Single-factor Pollution Index of Heavy Metals in the Surface Water of the coastal creeks of Ondo state

The surface water pollution status of

individual metal, is presented Table 6. Iron, Copper, Chromium and Cobalt revealed slight pollution (0.4 - 1.0). Zinc and Nickel showed medium pollution (1.0 - 2.0), while Lead and cadmium showed serious pollution (Pi>5.0**)** across the seasons.

Table 6:Single-factor pollution index of heavy metals in the surface water of the coastal creeks of Ondo State (Jun 2019 to April 2021).

Comprehensive Pollution Index (CPI) of heavy metals in the Surface Water of the coastal creeks of Ondo State

The comprehensive pollution index (CPI) of the coastal creeks of Ondo state is presented in Figure 2. The mean CPI values ranged from 3.34 at Ayetoro to 4.52 at Oroto. All the CPI mean values were greater than 2 (CPI>2), which indicated that the Coastal creeks of Ondo state were severely polluted across the sampling stations

Figure 2: The Comprehensive Pollution Index (CPI) of heavy metals in the surface water of the coastal creeks of Ondo State (Jun 2019 to April 2021).

Single-factor Pollution and Nemerrow Pollution of heavy metals in sediment of the coastal creeks of Ondo State

The values of single-factor pollution and Nemerrow pollution of heavy metals in sediment were presented in Table 7. The single-factor values of some metals (Zn, Pb and Cr) fell within the category of unpolluted sediment (Pij \leq 1), across stations and seasons. While other metals (Cu, Cd and Ni) were observed to have values that fell within the category of slight pollution (1 Pi \leq Pij \leq 2), across stations. However, the Nemerrow pollution values revealed that the sediment is slightly polluted at Ugbo, Ugbonla, Ayetoro and Oroto.

C ont a m ina t i on f a c t o r (C F) and contamination degree of Heavy metals in sediment of the coastal creeks of Ondo State

Contamination factor (CF) and Contamination degree (Cd) of Heavy metals in sediment of the coastal creeks of Ondo state are presented in Figure 3 and Figure 4 respectively. The values for contamination factor of all the metals revealed low pollution level of the sediment. However, the degree of contamination revealed a considerable level of pollution $(3 \leq Cf \leq 6)$ in the sediment across stations.

Geoaccumulation indices of heavy metals in the sediment of the coastal creeks of Ondo State.

The Geoaccumulation indices of heavy metals in sediment of the coastal creeks of Ondo state is presented in Table 8. The values are less than 0, which indicated unpolluted sediment (Igeo \leq 0). Except for Cd which ranged between slight pollution $(0 \leq \text{Igeo} < 1)$ to moderate pollution $(1 \leq \text{Igeo} \leq 2)$, across stations.

The single index of potential ecological risk and the comprehensive potential ecological risk of heavy metals in sediment of the coastal creeks of Ondo State.

The single index of potential ecological risk and the comprehensive potential ecological risk of heavy metals in of the coastal creeks of Ondo state are presented in Table 9. The values of the single index of potential ecological risk of all metals were less than 40 (Er <40), indicated low risk, except Cd that shows moderate risk $(40 \leq Er \leq 80)$ in the sediment of the coastal creeks of Ondo state. However, the comprehensive potential ecological risk showed low potential ecological risk as the values were greater than 150 (RI < 150), across stations.

Enrichment factor of heavy metals in sediment of the coastal creeks of Ondo State.

The enrichment factor of heavy metals in the sediment is presented in Figure 5. The values revealed that all the metals were extremely enriched in the sediment, since the value for each metal was greater than 40 (Er > 40).

Table 7: Single-factor pollution and Nemerow pollution of heavy metals in sediment of the coastal creeks of Ondo State

Figure 3: Contamination factors (CF) of heavy metals in sediment of the coastal creeks of Ondo State

Figure 4: Contamination degree (Cd) of heavy metals in sediment of the coastal creeks of Ondo State

പ്രപ്ര		Comprehensi ve potential ecological risk (RI)							
	Zn	Pb	Fe	Cu	\mathbf{C} r	C _d	Ni	∞	
Ugbo	0.20	0.04	0.00	2.12	0.09	55.00	1.18	4.35	62.97
Ugbonl a	0.30	0.05	0.00	2.16	0.06	57.00	1.21	4.52	65.29
Ayetoro	0.21	0.04	0.00	2.10	0.09	52.50	1.23	4.44	60.59
Oroto	0.29	0.05	0.00	2.18	0.06	59.50	1.34	4.80	68.21

Table 9: Ecological risk of heavy metals in sediment of the coastal creeks of Ondo State

Figure 5: Enrichment factor of heavy metals in sediment of the coastal creeks of Ondo State

Discussion

Many contaminants are released into the environment through myriads of anthropogenic activities within coastal communities (Onyema, 2009). Consequently, many tropical waters bordering the Atlantic Ocean are prone to pollution due to the fact that the region is densely populated, and highly industrialized. (Akanbi and Elenwo, 2015).

The concentration of heavy metals in surface

water and sediment of the coastal creeks of Ondo State were in the order; Fe>Zn>Cu> Ni> Co>Pb> Cd>Cr for surface water and Cu> Co> Ni >Zn >Fe> Pb > Cr> Cd for sediment. The recorded levels of most of the heavy metals were within WHO guidelines for the protection of surface water and sediment. However, Pb concentration was observed to be signicantly high in water which could be as a result of the prevailing crude oil exploration in the study area. Zinc levels in

surface water at the sampling stations exceeded FMEnv's permissible standards. This exceedance is likely due to steel production, coal burning, and the release of certain anthropogenic wastes into the environment. This finding further corroborated that of Mustapha et al. (2021), who inferred that coal burning and industrial activities in the Epe Lagoon led to a signicant increase in zinc levels in surface water.

The single-factor pollution index (Pi) of the surface water showed some levels of pollution of the waters that ranged from slight pollution to severe pollution, which could be due to a high level of anthropogenic activities along the coastal communities (Olawusi-Peters, 2021). However, the comprehensive pollution index (CPI) (which reflects the extent of overall pollution) of the study area revealed that the water was severely polluted with all the values greater than 2 (CPI <2) across stations and seasons. This is in accordance with the report of Iwegbue *et. al*. (2023), on the assessment of some physicochemical properties, metal concentrations, and water quality index from Bomadi Creek in the Niger Delta of Nigeria.

Metal concentration assessments in sediment revealed that some metals (Zn, Pb, and Cr) fell within the unpolluted category, while others (Cu, Cd, and Ni) exhibited slight pollution across stations. Nemerrow pollution values indicated slight to moderate pollution at different stations. This concurs with the findings of Ajani *et al.* (2021), that reported moderate pollution of sediment by heavy metals in Lagos lagoon.

Contamination factor values for all metals indicated low pollution levels in sediment, except for Cd in certain stations. The degree of contamination revealed a considerable level of pollution across stations and seasons, attributed to anthropogenic activities. This is consistent with findings of Popoola *et al*. (2015), who reported high contamination factor values for heavy metals in Lagos lagoon.

Geoaccumulation indices for individual heavy metals in sediment were mostly below one, indicating unpolluted sediment. Cd, however, showed slight to moderate pollution. The nding aligns with studies reporting slight to moderate pollution of Cd in coastal areas of the Egyptian Red Sea and is consistent with the findings of Mobarak *et al.* (2021).

Single indices of potential ecological risk for all metals were generally below 40, indicating low ecological risk, except for Cd, which suggested moderate risk in Mahin lagoon and its adjoining creeks. Comprehensive potential ecological risk also revealed low potential ecological risk overall, with values exceeding 150. This is in agreement with findings of Adesuyi *et a*l. (2021), who reported low potential ecological risk in Lagos lagoon wetlands. Enrichment factor analysis indicated that all metals were extremely enriched in sediment, with values exceeding 40. This finding is consistent with that of Nwineewii *et al*. (2018), who reported extremely high enrichment of heavy metals in sediment from New Calabar River, Nigeria.

Conclusion

Based on the results of this research, it is ascertained that coastal creeks of Ondo State are enriched and contaminated with heavy metals. The evaluated pollution indices revealed varying pollution status of the surface water and sediment from slight to severe pollution. In the sediment, the values of the single index of potential ecological risk of all metals indicated low risk, except Cd that revealed moderate risk $(40 \leq E r < 80)$. Meanwhile, the comprehensive potential ecological risk showed low potential ecological risk as the values were greater than 150 (RI < 150), across stations. Therefore, continuous investigations are necessary to forestall the potential health and ecological risk that may evolve as a result of continuous use of the water for domestic activities and consumption of fauna from the study area.

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