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RESEARCH ARTICLE

ASSESSMENT OF HEAVY METAL CONTAMINATION IN NTSINI RIVER SEDIMENTS FROM THE COASTAL PROTECTED AREA OF LIBREVILLE, NORTHWEST GABON

Norbert Ondo Zue Abaga[1], Stephane Mombo[2], Charly Mve Mfoumou[3] and Victory Nfoumou Obame[1]

1. Unite de Recherche en Sciences de la Terre et de l'Environnement (URESTE), Universite des Sciences et Techniques de Masuku (USTM), B.P. 913 Franceville, Gabon.
2. Unite de Recherche Agrobiologie, Universite des Sciences et Techniques de Masuku (USTM), B.P. 943 Franceville, Gabon.
3. Unite de Recherche en Chimie (URCHI), Universite des Sciences et Techniques de Masuku (USTM), B.P. 943 Franceville, Gabon.

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Abstract

An investigative study was conducted to determine the heavy metal contamination in sediments from the Ntsini River in the protected area Akanda National Park to Libreville, Gabon. A total of twelve sediment samples were collected from four locations (Ngwamba, Moka, Pages and Nendé). Sediments were analyzed for the six selected metals: Cd, Cr, Cu, Ni, Pb, and Zn using an Inductively Coupled Plasma-Atomic Emission Spectroscopy (ICP-AES). The high metal concentrations in the sediments were as follows: Zn > Cr > Ni > Pb > Cu > Cd. The highly significant positive Pearson correlations between the elements Cr, Cu, Ni, Pb and Zn indicate a common anthropogenic source of these metal elements. The CF shows an enrichment at all site for the metals Cd, Pb, and Zn. However, the CD indicates an exceptional Nendé station with moderate contamination for Cd. In this work, the results reveal low ecological risks (RI) for the metal Cr, Cu, Pb, Zn, and Ni in all stations, only Cd presents a moderate ecological risk in two stations: Ngwamba and Nendé. Therefore, effective mitigation strategies in response to metal contamination relevant to the Ntsini River in the Akanda National Park in Gabon are required to protect water quality and the health of local communities.

Introduction:-

The Republic of Gabon, located in central west coast of Africa, is traversed by the equator and bordered by Equatorial Guinea and Cameroon to the north, Republic of the Congo to the east and south, and 800 km of Atlantic Ocean coastline to the west [1]. Gabon, with Equatorial Guinea, Congo, Democratic Republic of Congo, Cameroon, and Central African Republic constitute the central Africa's tropical moist forests covering 1.8 million km², the second largest contiguous block in the word [2]. Gabon harbors around 80% of the country covered by moist

Corresponding Author:- Norbert Ondo Zue Abaga
Address:- Unite de Recherche en Sciences de la Terre et de l Environnement (URESTE), Universite des Sciences et Techniques de Masuku (USTM), B.P. 913 Franceville, Gabon.

tropical forest. This flora is classified in the Guineo-Congolian regional center of endemism [3], and its lowland diversity is among the richest in Africa [4]. In Gabon with surface area 267,667 km2 and a population of approximately 2.5 million inhabitants, 89% of the population lives in the two coastal urban areas of Libreville and Port-Gentil, a figure that is expected to rise up to 95% by 2050 [5]. The political and administrative capital, Libreville, is the most densely populated city with 3,700 inhabitants per km² [6]. As part of its policy to preserve and enhance natural ecosystems, particularly in the coastal zone, the Gabonese government has created three protected areas around Libreville that constitute the “Emerald Ark”, including Pongara National Park (PNP) to the south, Akanda National Park (ANP) to the north-east, and the coastal forest reserve of the Raponda Walker Arboretum (ARW) to the north [7]. These three areas are located between the Komo estuary and Mondah Bay include flooded forests and savannahs, herbaceous wetlands, mudflats, and mangrove forests [8]. These wetlands are internationally recognized for their importance to migratory shorebirds.

The ANP ecosystem consists mainly of mangrove trees [9] with six species of the Rhiziphoraceae, Avicenniaceae and Combretaceae families [10]. These trees species also provide valuables services, mainly by sustaining resource for local artisanal fisheries [11, 12], and contributing to carbon sequestration [13]. However, the proximity of ANP exposes them the increasing anthropic pressures due to population growth including cutting firewood [14], industrial and agricultural practices, domestic wastewater discharge, and traffic of fishing boats. Some work show a microbial water contamination by fecal indicator bacteria (FIB: *Escherichia coli* and *fecal streptococcus*) [15], and persistent organics pollutants and trace metals occurrence in marine organisms [16] in the Rivers located in ANP and connecting coastal Libreville and Monday Bay. Moreover, the work of Happi et al. [17] confirms a significant contamination of sediment by FIB and trace metals, with Cu, Pb in the estuarine sediments of ANP, and being associated with boat traffic and fuel spills. However, the specific metal contamination status of surface sediments in Ntsini River remains unavailable.

Because contaminants tend to accumulate in bottom sediments in aquatic systems, they are an important indicator of pollution [18]. Microorganisms, aquatic plants, and animals may bio-accumulate heavy metal residues in polluted environments. These organisms may then enter the human food chain and have long-term effects on ecosystems and human health [19, 20, and 21]. Sediments thus contaminated with trace metal elements constitute a reservoir and a potential source of contamination for water. Indeed, the slightest modification of environmental parameters, such as reactions accompanying natural or anthropogenic changes in redox potential or pH [22]; for example, the decrease in pH below a threshold defined for each metal causes its release [23] as well as the formation of complexes with organic or inorganic matter, can be easily mobilized [24, 25]. Knowing the state of contamination of sediments is an interesting decision-making tool because it allows for appropriate measures to be taken for sustainable management of an aquatic ecosystem [26].

Trace metal elements are considered dangerous pollutants because of their tendency to accumulate in biological organisms [27, 28] and to degrade the quality of ecosystems, thus threatening human health [29]. Due to its exposure to various human activities such as agriculture practices, industrial activities, and traffic of fishing boats, the Nstini River could have contaminated sediments acting as a reservoir and endogenous source of trace element pollution for water. The objective of the present study was an assessment of metal contamination levels in the sediments of Nstini River using for washing water supply, fishing, and irrigation sources. This study specifically aims to evaluate the concentration of sixth heavy metals, i.e., Cd, Cr, Cu, Ni, Pb, and Zn in the surface sediments in the Ntsini River of Emerald Ark in Gabon. The extent of ecological pollution due to these metals was characterized using the contamination factor (CF), contamination degree (CD), geo-accumulation index (Igeo), and potential ecological risk (RI) [30, 31]. This study aims to establish baseline data on heavy metal pollution in the sediments of Ntsini River, contributing to the future research and the development strategies for managing the protected areas and their associated aquatic ecosystem in the Republic of Gabon.

Material and Methods:-

Geographical location of the study Area:

This study was carried out in four sampling stations along the Ntsini River located in the Akanda National Park (ANP) at Libreville in Gabon (Figure 1). ANP covers 540 km², including 46% of estuarine and sea waters, and 33% of mangrove forests [32]. These mangroves are likely under stronger anthropogenic influences, including domestic wastewater discharge, fishing, agricultural and industrial activities. Ntsini River is located between latitudes 0°30N.and 0°36N, and longitudes 9°25E and 9°35E in the Northern part of Libreville [33, 34]. Study area enjoys equatorial wet climate with two dry seasons (May-September and December-January), and two wet seasons

(February-April and October-November), with the relative humidity around 70 to 80% throughout the year. The annual rainfall range is between 2,000 and 3,000 mm with an annual mean value of 2,500 mm rainfall per year, and a mean temperature of 25°C [35]. Due to regional high rainfall and its geographical location, Ntsini River is subject to two major phenomena: the runoff of anthropogenic wastes, and tidal fluctuations.

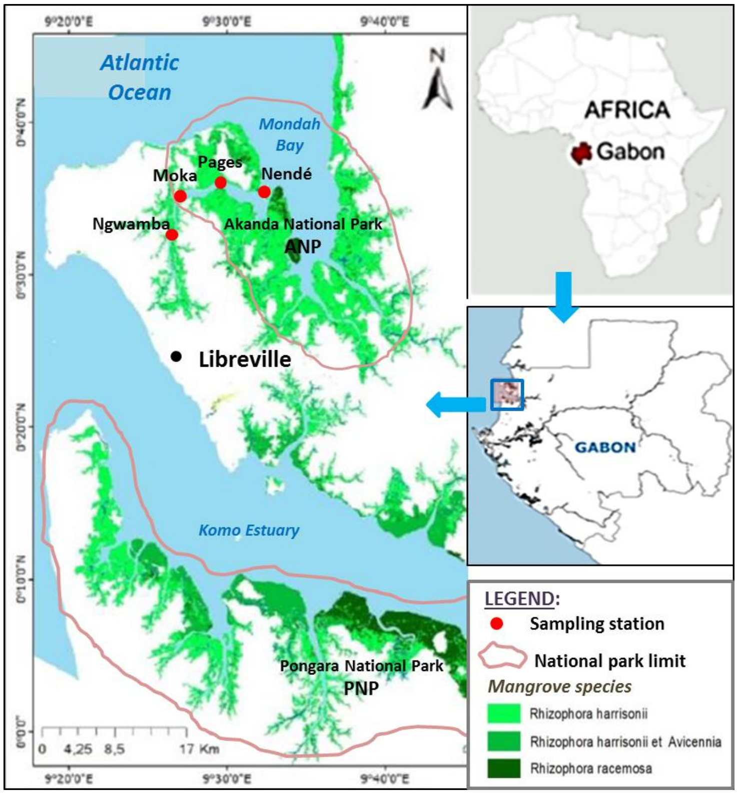


Figure 1: Location of sampling stations (in red points) in Akanda National Park (ANP) of the Emeraude Arc.

Collection and preparation of sediments samples:
Four selected stations sampling names Ngwamba, Moka, Pages and Nendé from upstream to downstream in figure 1 are presented in table 1. All sampling sites are located in the mudflats along the Nstisi River subject to the traffic of fishing boats. From a total of 4 sampling stations, 12 sediment samples were collected in November 2019 for the wet season. Three replicate sediments per station were sampled from the surface mudflats (0 -20 cm) using a cylindrical corer. Sediment samples were immediately stored in polyethylene bags and air dried at 25°C for 48 hours and sieved through a 2-mm sieve for further analysis.

Table 1: Location and description of sampling sites

Stations	Samples	Reason for choosing the station	GPS coordinates
Ngwamba	Ngwamba 1	Further upstream, mangrove with a high density of marsh tress. This station subjected to high anthropogenic discharges such as wastewater discharge, industrial and agricultural effluents, solid waste, oil spill	N 00°34'26.3'' E 009°27'16''
	Ngwamba 2		N 00°34'36.1'' E 009°27'39.8''
	Ngwamba 3		N 00°34'23.4'' E 009°27'15.3''
Moka	Moka 1	Intermediate station located on the bank of a tributary that flows into the Ntsini River, with a high density of marsh tress, moderate anthropogenic discharge such as agricultural effluents and oil spill from fishing boat traffic	N 00°35'34.4'' E 009°28'19.0''
	Moka 2		N 00°36'04.2'' E 009°28'13.8''
	Moka 3		N 00°36'18.2'' E 009°28'24.7''

<table>
<tr><td rowspan="3">Pages</td><td>Pages 1</td><td rowspan="3">Several small tributaries from agricultural areas flow into this canal. With a high density of marsh tress, agricultural effluents, and oil spill from fishing boat traffic</td><td>N 00°35’44.33’’
E 009°29’34.6’’</td></tr>
<tr><td>Pages 2</td><td>N 00°35’36.1’’
E 009°29’59.6’’</td></tr>
<tr><td>Pages 3</td><td>N 00°35’36.1’’
E 009°29’46.6’’</td></tr>
<tr><td rowspan="3">Nendé</td><td>Nendé 1</td><td rowspan="3">Located 13 km from the first station (Ngwamba), This station located towards Mondah Bay is the furthest downstream with low vegetation cover. It’s an islet used for monitoring the fishing boats.</td><td>N 00°35’55.3’’
E 009°32’45.9’’</td></tr>
<tr><td>Nendé 2</td><td>N 00°35’38.9’’
E 009°32’43.5’’</td></tr>
<tr><td>Nendé 3</td><td>N 00°36’01.9’’
E 009°32’35.5’’</td></tr>
</table>

Determining the pH, electrical conductivity and organic matter of sediment samples:
Sediment samples were dried for 6h at 60°C, sieved to obtain a 63 µm particle size, and homogenized for analyses. Sediment pH was measured in a 1:5 (w/v) sediment-to-water suspension using a multi-parameter (OAKTON instrumental). Organic matter (OM) content was determined by loss on ignition at 550°C for 4 hours [36]. The results are expressed as percentages on a dry weight basis.

Extraction of Metal from sediments:
The modified SERAS Method 3051A [37] was used to carry out the entire sediment digestion process in order to determine the heavy metal content. For each sediment, 0.5 g was digested in 9 mL of concentrated nitric acid (HNO_3), 3 mL of hydrofluoric acid (HF), and 2 mL of hydrochloric acid (HCl) using CEM microwave digestion system, MARS, with temperature control 180 °C for 10 minutes. The digested sediment was then filtered through 0.45-µm membrane filters and diluted to 50 mL with ultrapure water. The concentrations of six heavy metals - Cd, Cr, Cu, Pb, Ni, and Zn - were measured using a Thermo Scientific ICP-AES iCAP 6000 series. The method detection limits ($mg.kg^{-1}$) were 0.01 (Cd), 0.02 (Cr), 0.06 (Cu), 0.2 (Pb), 0.05 (Ni), and 0.2 (Zn). Sediment samples were analyzed in the three replicates, and results expressed as mean ± standard deviation (SD).

Calculation of pollution indices:
Several indicators such as the geo-accumulation index (I_{geo}), contamination factor (CF), contamination degree (CD), and potential ecological risk index (RI) were used for the first time in the Ntsini River to assess the status of river sediment pollution with potential toxic element. The selected indices are explained below.

Geo-accumulation Index (I_{geo}):-
The I_{geo} was proposed by Muller [38] to quantify pollution intensity in the environment using the following mathematical formula:

$$I_{geo} = \log_2\left(\frac{C_1}{1.5xC_b}\right)$$

Where C_i is the metal concentration in the sample, C_b is the UCC geochemical background concentration of metal [39], and 1.5 is the background matrix correction factor due the changes that may occur in lithology. The results are interpreted by Muller [38] as follows: $I_{geo} < 0$; unpolluted; $0 < I_{geo} \leq 1$: unpolluted to moderately polluted; $1 < I_{geo} \leq 2$: moderately polluted; $2 < I_{geo} \leq 3$: moderate to heavily polluted; $3 < I_{geo} \leq 4$: heavily polluted; $4 < I_{geo} \leq 5$: heavy to extremely polluted; and I_{geo} >: extremely polluted.

Contamination Factor (CF):-
The CF is an indicator often used to evaluate anthropogenic contributions and obtained by the following relationship:

$$CF = \frac{C_i \text{ (sediment sample)}}{C_r \text{ (Background)}}$$

Where C_i represents the concentration of an element in the sample and C_r the concentration in the sediment selected reference backgroud. Given that the predominant the bedrock in Akanda Park are limestone and sandstone, and since reference concentrations of heavy metals in aquatic sediments in the study area are currently unavailable, the average reference concentrations in sedimentary rocks determined by Rudnick and Gao [39] were used as geochemical background values. The CF values were interpreted according to Hakanson [40], where: CF < 1 indicates no enrichment; 1 < CF < 3 is moderate enrichment; 3 < CF < 6 is significant enrichment; and CF > 6 is very high enrichment.

Contamination degree (CD):-
The contamination degree (CD) proposed by Hakanson [40] can assess the overall contamination in a specific area, considering the sum of all the individual contamination factors (CFs). The CD index is calculated according to the following formula:

$$DC = \sum_{i}^{n} FC$$

Where n is the number of elements analyzed, FC is the concentration factor. The classification of CD values according to Hakanson [40] is as follows: CD < 8: low contamination; 8 < CD < 16: moderate contamination; 16 ≤ CD < 32: considerable contamination; and CD > 32: very high contamination indicating serious anthropogenic pollution.

Potential Ecological Risk Index (RI):-
This index is a mechanism for assessing the impact of potentially harmful components on aquatic behavior. The RI is obtained using following formula according to Hakanson [40] and Yi et al. [41]:

$$RI = \sum_{i=1}^{i} E\,r^{i} \qquad \text{avec } E_r^{i} = Tr \times FC$$

Where Er is the ecological risk factor for metal; Tr is the toxic response factor for a metal, which is defined for Cd = 30, Cr = 2, Cu = Pb = 5, Ni = 6, and Zn = 1 [42]. FC is the contamination factor. The ecological risk values are interpreted according to Hakanson [40], as shown in Table 2 below:

Table 2: Scale used to describe the risk factors of Er and RI [40] (1980).

Er	Potential ecological risk	RI	Ecological risk index
Er < 40	Low ecological risk	RI < 95	Low ecological risk
40 ≤ Er < 80	Moderate ecological risk	95 ≤ RI < 190	Moderate ecological risk
80 ≤ Er < 160	Considerable ecological risk	190 ≤ RI < 380	Considerable ecological risk
160 ≤ Er < 320	High ecological risk	380 ≤ RI	Very high ecological risk
320 ≤ Er	Very high ecological risk		

Statistical Analyses:
Analyses of variance (ANOVA) were performed at the four stations to test for significant differences in sediment parameters: organic matter, pH, electrical conductivity, and trace metal content. The spatial distribution and relationships between trace elements and sediment parameters at the different stations were examined using Principal Component Analysis (PCA). Relationships between the analyzed elements were tested using Pearson's coefficient with a threshold set at $p < 0.05$. Statistical analyses and graphs were performed using R software version 4.4.3.

Results:-

Physicochemical properties:
Values of organic matter (OM), pH and electrical conductivity (EC) obtained from sediments samples of Ntsini River are summarized in Table 3. The average amount of OM recorded for all sites exceeds 30% for the first three stations located upstream, following the order: Pages (33.31%) > Ngwamba (32.18%) > Moka (31.15%) (Table 3). The lowest mean of 24.45% is recorded for the Nendé station located furthest downstream, and is significantly lower ($p < 0.005$) than values obtained in the three Ngwamba, Moka, Pages stations. Mean pH values range from 5.53 to 7.13 (Table 3). Acidic pH values characterize stations more exposed to upstream anthropogenic pressures, such as Ngwamba (5.53), Moka (5.77), and Pages (5.47). Only the Nendé station present a neutral pH at 7.13. For mean pH values, the three upstream stations are very significantly lower ($p < 0.005$) than the Nendé station (Table 3). Concerning the electrical conductivity (EC), the results show an average variability from 7.64×10^3 to 10.23×10^3

μS.cm^{-1} for all sites studied (Table 3). Measurements recorded at the Ngwamba (9.38 × 10^3 μS.cm^{-1}), Moka (10.23 × 10^3 μS.cm^{-1}) and Pages (9.98 ×10^3 μS.cm^{-1}) stations are significantly higher ($p < 0.05$) than Nendé station (7.64 ×10^3 μS.cm^{-1}) (Table 3).

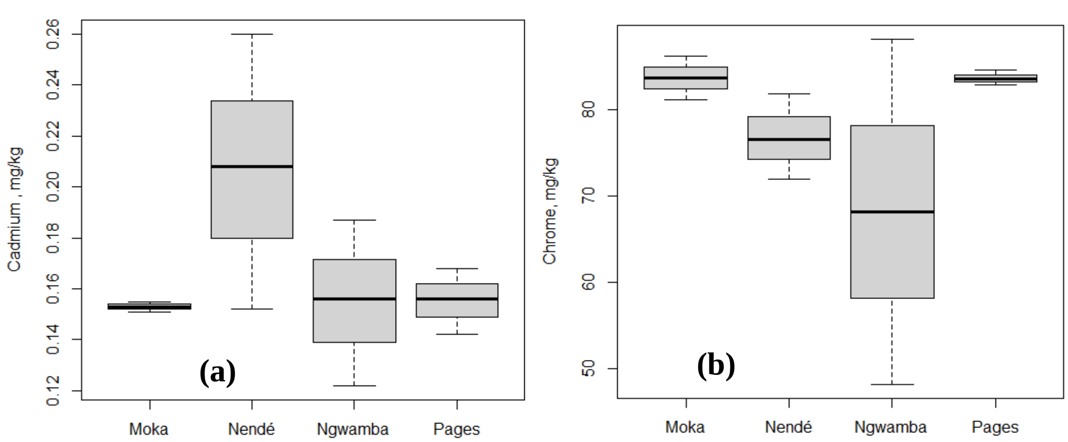
Table 3: Physicochemical parameters of sediments.

Sites		OM (%)	pH	EC (x10^3 μS.cm^{-1})
	minimum	29.91	5.45	9.36
Ngwamba	maximum	33.82	5.58	9.41
	mean	**32.18**	**5.53**	**9.38**
	s. deviation	2.029	0.07	0.02
	minimum	30.88	5.52	9.71
Moka	maximum	31.53	6.01	10.76
	mean	**31.15**	**5.77**	**10.23**
	s. deviation	0.34	0.24	0.52
	minimum	32.7	5.44	9.71
Pages	maximum	34.19	5.52	11.52
	mean	**33.31**	**5.47**	**9.98**
	s. deviation	0.77	0.04	1.53
	minimum	23.64	6.76	7.20
Nende	maximum	25.53	7.50	8.10
	mean	**24.45**	**7.13**	**7.64**
	s. deviation	0.34	0.37	0.45
ANOVA	F-value	32.87	35.98	5.79
	p-value	0.0000757***	0.0000542***	0.021*

OM : Organic Matter; EC : Electrical Conductivity ; p-value : probability value with *** p<0.001: Highly significant, **p<0.01: very significant, *p<0,05: significant.

Trace element contents in sediments:

Heavy metal concentrations in sediments are presented in figure 2. Metal concentrations ranged from: cadmium (Cd) 0.155 ± 0.03 to 0.207 ± 0.054 mg.kg^{-1}, chromium (Cr): 68.13 ± 19.95 to 83.63 ± 2.55 mg.kg^{-1}, copper (Cu) from 12.26 ± 2.99 to 14.95 ± 0.67 mg.kg^{-1}, nickel (Ni) from 20.50 ± 6.30 to 25.67 ± 0.35 mg.kg^{-1}, lead (Pb) from 15.8 ± 4.3 to 19.15 ± 1.12 mg.kg^{-1}, and zinc (Zn) from 53.33 ±16.60 to 88.63 ±21.75 mg.kg^{-1}of dry sediment. Higher metal concentrations were found at the following stations: Nendé (Cd = 0.21 mg.kg^{-1}), Ngwamba (Cr = 88.1 mg.kg^{-1}), Moka (Cu = 15.6 mg.kg^{-1}), Moka (Pb = 20.25 mg.kg^{-1}), Ngwamba (Ni = 26.8 mg.kg^{-1}), and Pages (Zn = 110.4 mg.kg^{-1}). However, ANOVA results do not show significant differences between the different stations for Cd (p = 0.196), Cr (p = 0.285), Cu (p = 0.205), Ni (p = 0.273), Pb (p = 0.294) and Zn (p = 0.066). This study determined the relative metal concentrations of sediment in the Ntsini River to be following order: Cr>Zn>Ni>Pb>Cu>Cd.



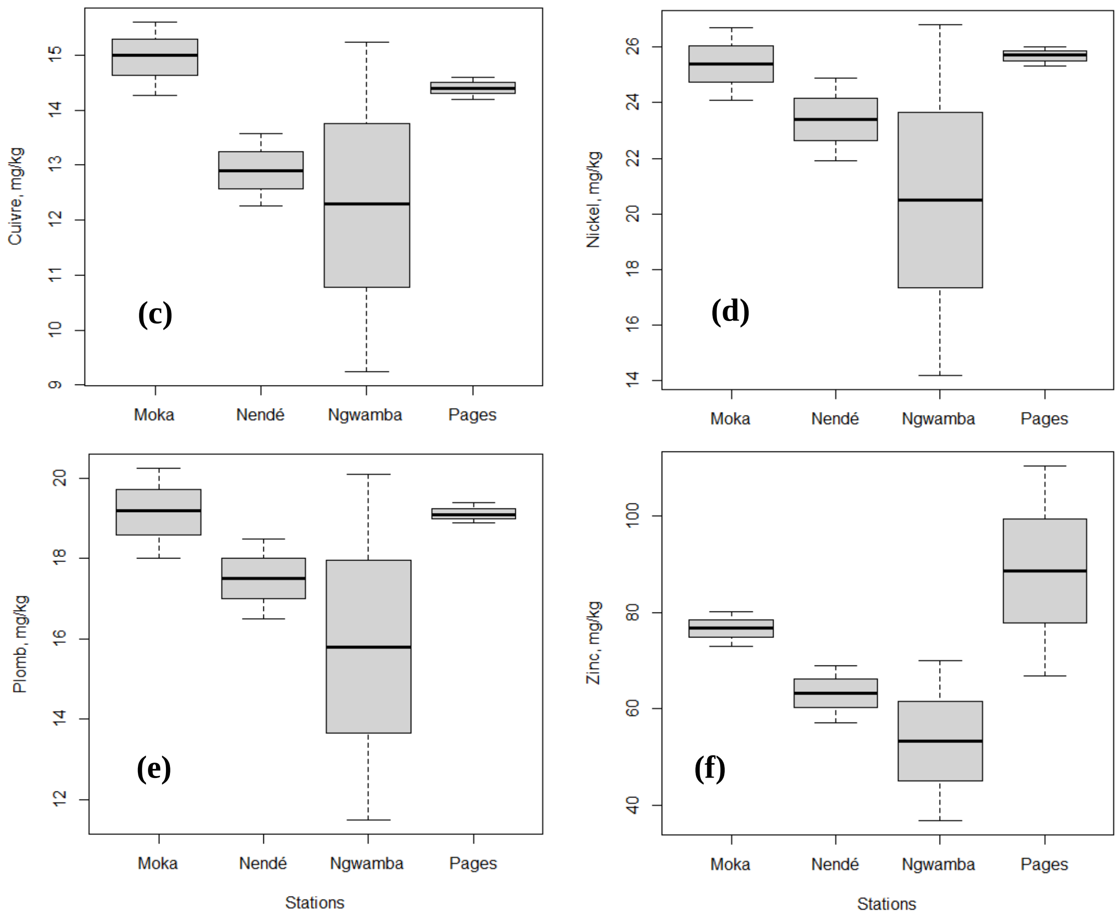


Figure 2: Heavy metal contents (en mg/kg) in the sediments of Ntsini River: Cd (a), Cr (b), Cu (c), Ni (d), Pb(e), and Zn (f).

Relationship between physicochemical parameters and trace metal elements:
Principal Component Analysis (PCA) analysis distinguished two significant factors, PC 1 and PC 2, which explain 50.51% and 30.58% of the original structure data, respectively (Figure 3 a, b). The total contribution of the distinguished factors is 81.09% with Cr, Cu, Ni, Pb and Zn concentrations were strongly positively correlated with PC1, while Cd concentrations were positively correlated with PC 2 (Figure 3 a). On the other hand, no correlation was observed between the distinguished factors PC1 and PC2 and the parameters OM, EC, and pH characterizing the physical and chemical properties of sediments from mudflats in Ntsini River. Indeed, the correlation matrix showed how the metals interacted with one another and with others parameters such as electrical conductivity (EC), organic matter content (OM) and pH (Table 4). Thus, Cr vs. Cu (r = 0.98), Cr vs. Ni (r = 0.99), Cr vs. Pb (r = 0.99), Cu vs. Ni (r = 0.98), Cu vs. Pb (r = 0.99), and Ni vs. Pb (r = 0.99) correlations showed a very strong positive relationship (Table 4). Zinc (Zn) vs. Cr (r = 0.66), Zn vs. Cu (r = 0.62), Zn vs. Ni (r = 0.65), and Zn vs. Pb (0.66) exhibited moderate positive relation (Table 4). However, Cd vs. Cr (r = 0.09), Cd vs. Cu (r = 0.908), Cd vs. Ni (r = 0.07), Cd vs. Pb (r = 0.09), and Cd vs. Zn (r = 0.03) correlations showed a non-existent relation. Therefore, no significant relationship was observed between the parameters pH, EC, MO and metals in the study.

In the Figure 3-b, the both two distinguished factors PC1 and PC2 highlight two groups of sediments in the Ntsini River: group 1 (G1) includes three stations Ngwamba, Moka and Page, carried by PC1 (50.51%), is especially characterized by low metal contamination by Cr, Cu, Ni, Pb and Zn, high OM values, higher EC values and acidic pH, and group 2 (G2), carried by PC2 (30.58%), and exclusively composed by the sediments form Nendé station, characterized by a neutral pH and a slightly higher Cd enrichment compared to the sediments of stations located further upstream river.

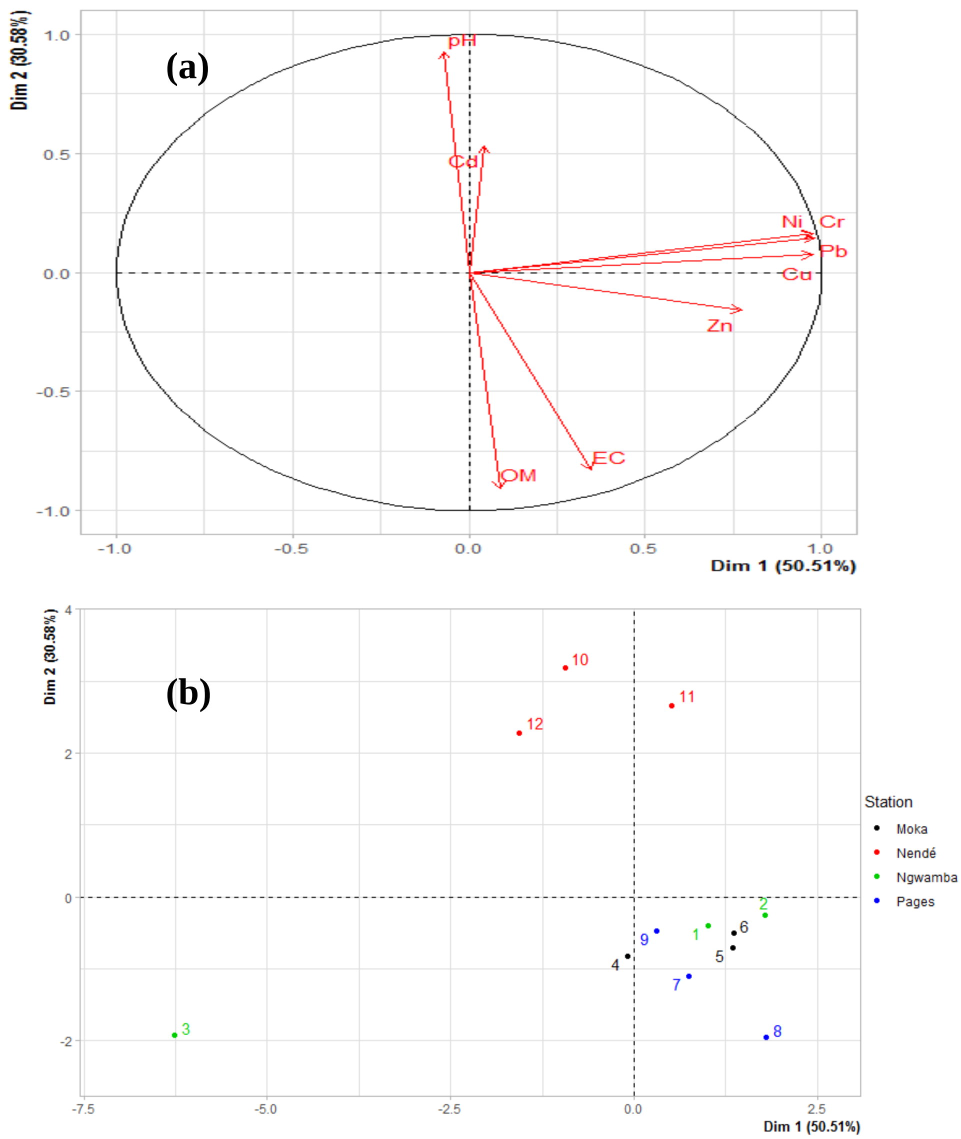


Figure 3: PCA characterization of the Ntsini River sediments based on: (a) physicochemical parameters and trace metal element concentrations; (b) the different sampling areas.

Table 4: Pearson correlation coefficients between parameters.

	Cd	**CE**	**Cr**	**Cu**	**MO**	**Ni**	**Pb**	**pH**
Cd								
CE	-0.40							
Cr	0.09	0.16						
Cu	0.08	0.21	0.98***					
MO	-0.22	0.69 *	-0.05	0.06				
Ni	0.07	0.16	0.99***	0.98***	-0.06			
Pb	0.09	0.17	0.99***	0.99***	-0.03	0.99***		
pH	0.38	-0.66*	0.05	-0.05	-0.93***	0.05	0.04	
Zn	0.03	0.58*	0.66*	0.62*	0.12	0.65*	0.66*	-0.06

****: Significant correlation at the 5% level (p<0.05);**
*****: Significant correlation at the 0,1% level (p<0.001)**

Assessment of the contamination degree of sediment:

The geo-accumulation index (I_{geo}) values (Table 5) of all samples for Cr, Zn, Ni, Pb, and Cu are less than 0 (I_{geo} < 0), indicating that the sediments are uncontaminated with these elements. As for Cd, its average values (0 < I_{geo} < 1) reveal slight Cd contamination in all analyzed sediments in this study [38]. The contamination factors (CF) and the contamination degree (DC) values are summarized in Table 6. The CF values less than 1 for Cr, Cu, and Pb indicate that these sediments are uncontaminated with these three metal elements. However, for all samples, the CF values for Zn range from 0.22 to 2.07; those for Pb from 0.35 to 1.25 and the CF values for Cd from 1.91 to 5.41. All sediments show moderate contamination with Zn and Pb (1 < Igeo < 3) [40]. For Cd, there is moderate contamination (1<CF<3) in sediments from Moka and Pages stations and some sediments from Ngwamba and Nendé stations, and significant contamination (3<CF<6) of at least one sampling site from Ngwamba (CF = 3.70) and Nendé (CF = 5.41) stations. The contamination degree (CD) values confirm that the sediments at the Moka and Pages stations have a low degree of contamination in Cr, Zn, Ni, Pb and Cu (CD < 8), while for the Ngwamba and Nendé stations, 1/3 of the sampling sites have an 8<CD<16 indicating moderate contamination, particularly in Cd. This Cd contamination of the sediments appears to be more significant for the Nendé station located furthest downstream of the Ntsini River, towards Mondah Bay.

Table 5: Geo-accumulation index (I_{geo}) of the metal elements in sediments.

Sites	Geo-accumulation index (I_{geo})					
	Cr	Zn	Ni	Pb	Cu	Cd
Ngwamba	- 0.30 ± 0.36	- 0.38 ± 0.39	- 0.48 ± 0.38	- 0.22 ± 0.31	- 0.55 ± 0.27	0.21 ± 0.16
Moka	- 0.14 ± 0.03	- 0.13 ± 0.04	- 0.30 ± 0.04	- 0.07 ± 0.05	- 0.42 ± 0.04	0.22 ± 0.01
Pages	- 0.14 ± 0.01	- 0.10 ± 0.20	- 0.29 ± 0.01	- 0.08 ± 0.01	- 0.43 ± 0.01	0.23 ± 0.05
Nende	- 0.18 ± 0.05	- 0.22 ± 0.08	- 0.34 ± 0.06	- 0.11 ± 0.05	- 0.48 ± 0.04	0.32 ± 0.21
***UCC**	76	69.5	33.5	15.12	26	0.061

***UCC: Reference values of the Earth's crust in mg/kg [39].**

Table 6: Contamination factors (CF) and contamination degrees (CD) of sediments witgh metallic trace elements.

Sites	Contamination factor (CF)						$CD = \sum CF_i$
	Cr	Zn	Ni	Pb	Cu	Cd	
Ngwamba	0.74 ± 0.43	0.79 ± 0.49	0.44 ± 0.27	0.93 ± 0.50	0.44 ± 0.21	2.56 ± 0.99	5.90 ± 2.55
Moka	0.91 ± 0.06	1.15 ± 0.11	0.54 ± 0.05	1.13 ± 0.13	0.54 ± 0.05	2.50 ± 0.07	6.76 ± 0.39
Pages	0.91 ± 0.01	1.32 ± 0.65	0.55 ± 0.01	1.12 ± 0.02	0.51 ± 0.01	2.55 ± 0.31	6.97 ± 0.60
Nende	0.83 ± 0.10	0.94 ± 0.17	0.50 ± 0.07	1.03 ± 0.11	0.46 ± 0.04	3.41 ± 1.75	7.17 ± 1.68

The potential risk factor (Er) and ecological risk index (RI) values are summarized in Table 7. The results show that the Er values are much lower than 40 (Er<40) for the elements Cr (from 0.4 to 2.1). Zn (from 0.7 to 2.1). Ni (from 0.8 to 3.7). Pb (from 1.8 to 6.3) and Cu (from 1.0 to 2.9), which indicates that the potential ecological risk for these elements is low according to Hakanson [40] (Table 2). In contrast, the Er values in the sediments of the Ngwamba (from 57.3 to 111.0), Pages (from 72.9 to 77.4), Pages (from 69.2 to 87.3) and Nendé (from 65.4 to 162.4) stations potentially imply moderate ecological risks for Cd (40 ≤ Er < 80) for the sediments of the Moka and Pages stations, and potentially considerable ecological risks for Cd (80 ≤ Er < 160) for some sediments of the Ngwamba and Nendé stations. The RI values indicate that there is an existing ecological risk for the element Cd that is moderate (95 ≤ RI < 190) for the Ngwamba and Nendé stations [40].

Table 7: Potential risk factor (Er) and ecological risk index (RI) of trace elements in sediments.

Sites	Potential risk factor (Er)						RI
	Cr	Zn	Ni	Pb	Cu	Cd	
Ngwamba	1.48 ± 0.87	0.79 ± 0.49	2.62 ± 1.61	4.65 ± 2.52	2.19 ± 1.06	76.91 ± 29.68	88.65 ± 33.40
Moka	1.82 ± 0.12	1.15 ± 0.11	3.25 ± 0.31	5.64 ± 0.64	2.68 ± 0.24	75.12 ± 2.2	89.65 ± 3.38
Pages	1.82 ± 0.03	1.32 ± 0.65	3.28 ± 0.08	5.61 ± 0.09	2.57± 0.06	76.63 ± 9.47	91.23 ± 9.10
Nende	1.66 ± 0.21	0.94 ± 0.17	2.98 ± 039	5.15 ± 0.55	2.30 ± 0.23	102.29 ± 52.50	115.34 ± 51.85

Discussion:-

Physicochemical properties and heavy metals are influenced by variations of vegetal configuration, land runoff, and anthropogenic activities. The ACP highlight two types of sediments in the Nstini River: the first sediment type moderately acidic located in three upstream stations Ngwamba, Moka and Pages, characterized by low metal contamination by Cr, Cu, Ni, Pb and Zn, and the high values of OM and EC, and the second sediments located in dowstream Nendé station characterized by neutral pH and a few Cd enrichment. Organic matter content show a very strong negative correlation with pH (r=-0.93), possibly linked to the release of H^+ protons during the organic matter decomposition process, which would increase sediment acidity. Conversely, high H^+ ion saturation would explain the EC increasing, and the positive correlation between OM vs. EC (r = 0.66) (Table 4). Thus, EC values in all sediments are significantly higher compared to the OMS standard value (2500 µS/cm) [43]. In this study, no significant relationship between the parameters pH, EC, MO and metals element indicates a natural source for OM, which come from the natural decoposition of plants and animals in the mangrove, and the mixture sources of ETM linked to the pedogeochemical background on the one hand, and from anthropogenic activities on the other. An exceptionally low positive relationship has been found for EC vs. Zn (r = 0.58) confirm that Zinc (Zn) metal tends to associate with mud and organic debris [44].

The observed relationships between Cr, vs. Cu, Ni, Pb, and Zn, are very high (r ≥ 0.98). For the six studied metals, Cd is an isolated case and shows no relationship with all other metals (r values from 0.03 (Zn) to 0.09 (Cr, Pb). The Cd source would be completely different from that of Cr, Cu, Ni, Pb, and Zn. The highest Cd concentrations were found downstream of the Nstini River (Nendé station) at sites wich are directly connected to Mondah Bay with less human activities such as domestic and industrial drainage, agricultural practice, and wastewater discharges, suggesting Cd source may be mostly natural. An acidic pH in upstream stations (Ngwamba, Moka and Pages) would promote the trace elements solubilization in the sediments of the first three stations while a neutral pH in downstream station (Nendé) would promote Cd adsorption on the sediments [45]. According to the metal concentrations in this investigation were as follows: Zn>Cr>Ni>Pb>Cu>Cd. Very high relationships observed for Cr and others metal (Cu, Ni, Pb, and Zn) indicated metal contamination of Ntsini River linked to both natural source such as weathering of rocks, and anthropogenic activities, including wastewater discharges, industrial and agricultural practices, and traffic of fishing boats. These data emphasize that heavy metal contamination of sediments in coastal area of Libreville is notably associated with boat traffic and fuel spills [17].

Comparison of heavy metal in the sediments of Ntsini River with other selected Sediment Rivers:

A comparison of the heavy metal concentration in sediments to other selected rivers is shown in Table 8. The concentrations of Cr and Pb in Moka, Zn in Pages and Cd in Nendé were higher than the geochemical background of the UCC continental crust [39]. Remarkably, Cr and Ni concentrations observed in this study exceeded the standards established by the World Health Organization (WHO). However, the concentrations levels of Cd, Cu, Pb, and Zn in the surface sediments of the Ntsini River were found to be within the recommended criteria set by the World Health Organization (WHO). In this study, higher Cr and Ni average values were especially obtained from the upstream station (Ngwamba). Metal enrichment of upstream sediments of Ntsini River appears to be linked to anthropogenic sources such as fishing boat traffic, waste incineration, and fertilizers used in agriculture [46, 47]. The concentrations of Cr, Cu, Pb, and Zn observed in the present study were comparatively lower than the values reported in previous studies conducted on the coastal rivers and wetland zones of Libreville in Gabon [17].

Generally, the concentrations of Pb, Ni, Cd, and Zn in sediments from the marine protected areas of Libreville, including Ntsini River, are comparatively higher than metal contents obtained in the Sediment Rivers from Nigeria

[48], Brazil [49] and Thailand [50]. In comparison with the Atlantic coast of DR Congo [51] which shows high metal concentrations in sediments exceeding the standards established by the WHO, our results show that only Cr and Ni concentrations exceeded these WHO Standards [50] (table 8). Our results indicate a low level of metal contamination in the sediments from Ntsini River, with a significant ecological risk linked to high Cd contamination, particularly in the sediments located at downstream station. In this study, the results highlight the importance of monitoring and regulating human activities in ANP to minimize environmental contamination.

Table 8: Comparative analysis of heavy metal concentration in marine and coastal sediments: a global perspective including national and international studies.

Study area	Mean Concentration of Heavy metal in the sediment ($mg.kg^{-1}$)						Country	Reference
	Cd	Cr	Cu	Pb	Ni	Zn		
Nstini River	0.17	78.02	13.63	17.89	23.74	70.42	Gabon	This study
Wetlands in ANP	0.13	101.59	21.44	28.00	na	114.83	Gabon	[17]
coastal area of Libreville	0.13	57.26	23.79	41.91	na	142.09	Gabon	[17]
Atlantic Coast	na	187	217	125	30	na	DR Congo	[51]
Ondo coastal area (Awoye)	na	0.92	3.21	14.5	6.69	7.27	Nigeria	[48]
Tropical mangrove areas	0.06	61.2	12.14	15.08		45.00	Brazil	[49]
*WHO	0.99	43.4	31.6	35.8	22.7	121.0		[50]
**UCC background level	0.061	76	26	15.12	33.5	69.5		[39]

***WHO: Word Health Organization, DR Congo: Democratic Republic of Congo**
****UCC: Reference values of the Earth's crust in mg/kg [39]**

Values of contamination factor (CF) and degree of contamination (DC) confirm a sediment contamination with heavy metals following the order: Cd>Zn>Pb>Cr>Ni>Cu, and the most contaminated sediments are classified in the stations according to the order Nendé>Ngwamba>Pages>Moka. These metals discharged upstream would be redistributed along the river by the tidal current which is favored by a moderately high flow (74 m^3/s) of the Mondah Bay [52], which would explain a low degree of contamination of the sediments (DC < 8) in Cr, Cu, Ni, Pb, Zn and moderate (8<DC<16) in Cd along the Ntsini channel [40]. For all trace elements, only Cd potentially presents a moderate ecological risk ($40 \leq Er < 80$) in sediments for the Moka and Pages stations, and a considerable ecological risk for the Nendé and Ngwamba stations. However, the highest ecological risk index (RI) value for Cd obtained in sediments from the Nendé station (RI = 174.9) shows that these sediments present a greater ecological risk compared to sediments from other stations. Cadmium (Cd) is the only element that presents a significant ecological risk. It is a bioaccumulative trace element with high toxicity in living organisms [53, 54]. Trace elements that are adsorbed on sediments can be released at the slightest modification of environmental parameters such as changes in redox potential, salinity or pH, thus causing their concentration in the water column [22, 23]. In the coastal zone, Cd founded in dissolved form and associated with chloride ions [55], is bioavailable to fish species, which could have deleterious effects on aquatic life and the health of consumer peoples of fish from the Ntsini River. This results confirms the need for regular monitoring of the human activities in coastal zone to avoid further contamination by Cd, Cr and Ni and subsequent pollution in the ecosystems of Ntsini River.

Conclusion:-

In this work, the contamination status of the surface sediment samples along Ntsini River were analyzed with heavy metal contamination and multiple sediment quality indices, in Akanda National Park in Libreville, Northwestern Gabon. In this protected estuarine area, there are two types of sediments from four different stations: sediments from the Ngwamba, Moka, and Pages mangrove mudflat stations characterized by an acidic pH, a high amount of organic matter, and similar Cr, Cu, Ni, Pb, and Zn contents, and sediments further downstream from the Nendé station, characterized by a neutral pH, a lower amount of organic matter, and especially higher Cd contents. The results showed Cd contamination in all sediments, and Cr, Pb, and Zn in the sediments from the Moka and Pages stations, their concentrations being higher than those of the geochemical background of the continental crust. The highly significant positive Pearson correlations ($p < 0.001$) between the elements Cr, Cu, Ni, Pb and Zn indicate a common anthropogenic source of these metal elements. In this study, the contamination factors (CF) and the degree of contamination (DC) respectively reveal the contamination status of sediments by the elements following the order Cd > Zn > Pb > Cr > Ni > Cu, and the ranking of stations according to their overall contamination state: Nendé > Ngwamba > Pages > Moka. However, the element Cd was determined as the only one to have significantly contaminated the sediments of the Ntsini ($0 < I_{géo} < 1$) and to have caused a moderate ecological risk ($40 \leq Er < 80$) for the Moka and Pages stations, and considerable ($40 \leq Er < 80$) for the sediments of the Nendé and Ngwamba stations. The values of the ecological risk index (RI) confirm an ecological risk for Cd element which is moderate for the sediments of the Ngwamba and Nendé stations ($95 \leq RI < 190$). A preventative policing system should be established to regulate potentially polluting activities commonly carried out in the coastal area of Libreville, such as waste and wastewater discharges, runoff of agricultural and industrial effluents, and boat fishing traffic.

Conflits of interest

The authors do not have any possible conflicts of interest.

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