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The effect of pH on heavy metals availability on crude oil remediated and non-remediated soil, Nkeleoken Community Eleme, Rivers State, Nigeria

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Abstract

This study was aimed at examining the influence of pH on heavy metals availability in polluted and non-polluted soils. The findings showed that metal availability depends on soil solution which is pH dependent. The values of pH in Nkeleoken remediated soil (RSENa, RSENb, RSENc) in dry season were 6.01, 5.87, 5.78 and 6.64, 5.77, 5.68 in wet season with mean values of 5.89±0.05 and 6.03±0.53 while the pH of the contaminated soil ranged between 4.31 – 4.41. The mean values of heavy metals in Nkeleoken remediated soil are; Se (318±9.88; 326±11.6 mg/kg), Cd (3.65; 3.82±1.09 mg/kg), V (1095±71.05; 1189±149 mg/kg), Ba (8.98±1.53; 9.40±1.89 mg/kg), Cu (8.19±0.08; 8.84±0.14 mg/kg), Ni (0.02±0.01 mg/kg), As (5.52±2.35; 10.3±2.38mg/kg), Pb (1.73±0.32; 2.70±1.38 mg/kg) and Cr (23.1±4.05; 23.1±4.05 mg/kg). The results revealed that V and Se had the highest concentration level followed by As while the least concentrations were observed in Cd, Ba, Pb and Cr in both dry and wet season. The concentrations of heavy metals in the non-remediated soil ranged from 866 – 868 mg/kg, 1356 – 6332 mg/kg, 211 – 218 mg/kg, 73.6 – 79.0, and 866 – 868 mg/kg for Se, V, Ba Pb and as in dry and wet seasons. However, Cr concentrations ranged from 6.03 – 7.02 mg/kg. The concentrations of V and Se in the non-remediated soils (NRSENx - NRSENz) during dry and wet seasons were higher than their corresponding in the remediated soils likewise the recommended values set by DPR (2012).

Keywords: Remediation; Soil; pH; Pollution; Availability; Solubility

1 Introduction

Environmental pollution with toxic heavy metals is now a worldwide Issue. As a result of the increasing concern on the impending effects of the metallic pollutants on the environment and human health, the research on fundamental, applied and health facets of heavy metals in the environment is also increasing [1]. Industrial processes that discharge a variety of heavy metals into waterways include mining, smelting and oil refining [2]. Large quantities of these heavy metals are released into soil as a result of increased anthropogenic activities such as agricultural practices, waste disposal and petrochemical activities leading to the pollution of the soil [3]. Soil is a natural reservoir of metals whose concentrations are related with several factors such as biological and biogeochemical cycling, parent material and mineralogy, organic matter, soil pH and microbial activities [4]. Different mechanisms are accountable for the adsorption and retention of heavy metals in polluted soils: specific adsorption, cation exchange, organic complexation and co-precipitation [5]. The distribution coefficient of heavy metals differs due to pH, redox conditions, ionic strength and dissolved organic matter concentrations.

The remediation of soils refers to practices of either removing contaminants or converting them into less mobile species; that is, into less bioavailable forms. The selection of a method is generally based on the nature of the contaminants, the

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soil type, and the characteristics of the contaminated site. Methods for remediating metal-polluted soils have been widely investigated and discussed [6, 7].

2 Material and methods

2.1 Description of study area

2.1.1 Eleme

This is one of the local government area of Rivers state. It is one of the settlement with refinery in Ogoniland. It lies between Latitude 4.7874 and longitude 7.1433: The sampling locations covers two community (Alode and Nkeleoken).

- The Non-remediated site is an SPDC Well in Alode-Ogale with UNEP CODE: 005 – 001. The coordinates of the sample locations are (E295804, N2955341); (E295758, N533914).
- The Nkeleoken remediated site is an NNPC and SPDC Trunk lines with UNEP Reference Code: 002 – 002. The sampling coordinates are (E291066, N527226); (E291075, N527217); (E291039, N527162).

2.2 Collection and preparation of soil samples

Stratified and systematic sampling procedure was applied for soil sample collection according to [8] with slight alteration. Under this sampling procedure, the remediated site, Nkeleoken in Eleme was broken into three sub-populations labelled a, b and c. The non-remediated site Alode was also divided as x, y and z. Samples were taken randomly in individual sub-population at a distance of 150 meters from each other to enable detailed study on individual sub-population and increases the precision and accuracy of the estimate over the entire sites. In each sub-population, approximately 10 – 20 g soil samples were randomly collected at the depth of 0 –15 cm for remediated soil and 3 m for non-remediated soil with manual auger. The samples were thoroughly mixed to obtain representative samples which was manually sorted to eliminate pebbles and coarse materials, and air-dry at room temperature over three days with occasional breaking of aggregated materials with wooden roller. This was follow by sieving through a nonmetallic sieve of mesh size 2 mm diameter to retain bigger size soil particles. The air dried 2 mm soil samples were stored in glass bottle at room temperature for extraction and analysis. Samples were collected in the month of March and September, 2021.

2.3 Determination of Soil pH

Hydrogen ion concentration was determined by using a method described by McLean adopted from [9]. In this method, 1:10 ratio of soil to distilled water mixture was prepared by weighing 10 g of fine sieved soil into a glass beaker followed by addition of 100 mL of de-ionized water which was stirred gently to enhance H^+ (Hydrogen ions) release from the soil. The resulting mixtures was allowed to stand for 30 min, then the soil pH was determined by inserting the pH probe into the mixture.

2.4 Determination of total metal concentration in soil.

A 1.0 g of the soil was weighed into a kjeldahl flask and digested with 25 mL of aqua regia (1:3 HNO_3 /HCl v/v) at 90 °C for 1 hr, 30 minutes on heating mantle during which complete decomposition of soils was achieved. The digest was allowed to cool to room temperature. To the digests was added 10 mL deionized water. The digest was decanted into a 50 mL polyethylene bottle and was made up to mark with deionized water [10]. It was stored for further analysis by Microplasma Atomic Emission Spectroscopy (MP-AES, Agilent 4210).

2.5 Statistical Analysis

Data was subjected to statistical analysis of significance by using the one way analysis of variance (ANOVA) to assess significant disparity in the concentration levels of the heavy metals in the soils sample across the sampling sites and seasons. Probability less than 0.05 ($p<0.05$) was considered statistically significant.

3 Results and discussion

The results of the degree of acidity and alkalinity of the soil presented in Table 1 shows that, the values of pH in Nkeleoken remediated soil ($RSEN_a$, $RSEN_b$, $RSEN_c$) in dry season were 6.01, 5.87, 5.78 and 6.64, 5.77, 5.68 in wet season with mean values of 5.89±0.05 and 6.03±0.53 while the pH of the non-remediated soil (NRSENx - NRSENz) ranged between 4.31 – 4.41 with mean value of 4.37±0.05 in the dry season and in the wet season the values decreases as follows; 4.30, 4.20 and 4.25 with mean value of 4.25±0.05 across $NRSEA_x$, $NRSEA_y$ and $NRSEA_z$.

From the results it is noticeable that the pH levels of the remediated and non-remediated soils in dry season were lower than those obtained in the wet season and in all the sample locations. Similar pH results (4.70 – 5.60) was reported by [11] in their findings on post-impact assessment of oil pollution on some soil characteristics in Ikot Abasi, Niger Delta Region, Nigeria

The reason for low soil pH values in dry season was due to the presence of organic compounds containing hydrogen that decomposes and released H^+ onto the soil. Hence, the pH decreases results in an increase of metal concentrations in the soil [12]. The values of pH obtained in this study is in agreement with the findings of [13] which recorded pH values ranged from 5.54 – 5.59 respectively.

Table 1 Results of Soil pH in Nkeleoken Remediated and Non-remediated Sites Eleme during dry and wet season

Sample Locations/Seasons	Dry Season	Wet Season
RSENa	6.010	6.640
RSENb	5.870	5.770
RSENc	5.780	5.680
Mean ± SD	5.89±0.05	5.820±0.530
NRSEAa	4.400	4.300
NRSEAb	4.310	4.200
NRSEAc	4.410	4.250
Mean±SD	4.370±0.050	4.250±0.050

Despite the remediation work conducted on the impacted soil, it still recorded low pH values, though above their corresponding non-remediated soils. However, the non-remediated soil recorded lower pH values which means they are more acidic than the remediated soil. This implies that the remediation work had raise the soil pH to small extent but still below the recommended pH values for agricultural soil. This implies that the soils are highly acidic and may have high tendency of increasing heavy metals and its mobility [12].

Raymond and Yuwaree [14] stated in their work that, a change in the soil pH may results in a corresponding change of the dominant retention mechanism of heavy metals in the soils. High concentrations of heavy metals are available in the soils if the soil buffer capacity remains low such that it cannot resist a change in pH. The selectivity order of heavy metal retention in soils depends on the pH of soil solution. At soil solution pH values above 4 or 5, when precipitation prevails, the selectivity order obtained is given as Pb > Cu ≥ Cd and at lower soil solution pH values, the selectivity order obtained is given as Pb > Cd > Cu.

The results of the heavy metals concentrations in dry and wet seasons were presented in table 2.

Nkeleoken remediated soil (RSENa – $RSEN_c$) had mean values; Se (318±9.88; 326±11.6 mg/kg), Cd (3.65; 3.82±1.09 mg/kg), V (1095±71.05; 1189±149 mg/kg), Ba (8.98±1.53; 9.40±1.89 mg/kg), Cu (8.19±0.08; 8.84±0.14 mg/kg), Ni (0.02±0.01 mg/kg), As (5.52±2.35; 10.3±2.38mg/kg), Pb (1.73±0.32; 2.70±1.38 mg/kg) and Cr (23.1±4.05; 23.1±4.05 mg/kg). The mean values of heavy metals obtained in Nkeleoken remediated soil indicates that V and Se had the highest concentration level followed by As while the least concentrations were found in Cd, Ba, Pb and Cr in both dry and wet season respectively.

However, the mean concentrations of Se and V in dry and wet seasons which were above the acceptable values by [15, 16] for metals in soil. Thus, this findings suggests that the studied locations were severely contaminated by the referred heavy metals. The high concentration level of Se and V could be attributed to the nature of the parent material of the soils and this results agreed with the findings of David and Minati [17].

Table 2 Heavy metals concentrations (mg/kg) in soil from Nkeleoken remediated and non-remediated Soil Eleme in dry season

Samples	Se	Cd	V	Ba	Cu	Ni	As	Pb	Cr
RSENa	314±0.80	2.62±0.25	1017±2.80	11.5±0.85	8.93±0.85	ND	7.87±1.60	3.14±0.80	2.62±0.25
RSENb	324±0.30	3.57±1.45	1382±3.80	9.86±0.75	8.95±0.55	ND	9.41±1.45	3.24±0.30	3.57±1.45
RSENc	342±0.70	5.27±1.35	1168±2.20	6.91±0.25	8.64±0.95	ND	13.5±1.65	3.42±0.70	5.27±1.35
NRSEAx	866±0.20	6.04±0.15	6332±1.20	218±0.54	50.1±0.85	9.28±0.25	79.0±0.15	8.66±0.20	6.04±0.15
NRSEAy	867±0.20	6.13±0.15	2928±2.20	211±0.66	50.4±0.85	8.35±0.25	77.1±0.15	8.67±0.20	6.13±0.15
NRSEAz	868±0.10	7.02±0.15	1356±1.20	211±0.50	50.2±0.85	7.25±0.27	73.6±0.15	8.68±0.10	7.02±0.15
DPR (2012)	**28.0**	**0.80**	**0.50**	**200**	**36.0**	**35.0**	**29.0**	**85.0**	**100**

ND: Not Detected, SD: Standard deviation

Organic complexes of Cu and Pb remain stable until pH 4, this leads to low concentration of Cu and Pb while complexes of Cd is less stable and dissociate when the pH is below 6 and this led to high levels of Cd. The low concentrations of Cr in the remediated soil was due to moderately high pH level as compared to the non-remediated soil. This is supported by previous research of [18] that low solubility of Cr and strong retention on soil particle surfaces limits its availability in soils with pH within 5 - 6.

The concentrations of heavy metals in the non-remediated soil ranged from 866 – 868 mg/kg, 1356 – 6332 mg/kg, 211 – 218 mg/kg, 73.6 – 79.0 mg/kg, 866 – 868 mg/kg for Se, V, Ba Pb and As in dry and wet seasons. However, Cr concentrations ranged from 6.03 – 7.02 mg/kg. The concentrations of V and Se in the non-remediated soils during dry and wet seasons were higher than their corresponding in the remediates soils likewise the recommended values set by DPR [15] while Ba and Cu recorded higher values than the permissible limit only in the non-remediated soils.

Marlena *et al.,* [19] reported that Vanadium availability and uptake of V depends on soil pH, increased vanadium uptake was observed at low pH (4 - 5) and uptake in the range of 5 - 8 was lower but stabilized. High pH (alkaline) hindered vanadium uptake in roots.

Table 3 Heavy metals concentrations (mg/kg) in soil from Nkeleoken remediated and non-remediated Soil Eleme in wet season

Samples	Se	Cd	V	Ba	Cu	Ni	As	Pb	Cr
RSENa	309±1.30	2.52±0.15	1009±5.20	10.5±0.85	8.06±0.15	0.02±0.05	7.57±0.95	3.14±0.85	2.07±045
RSENb	314±1.70	3.47±0.15	1183±5.20	9.56±0.55	8.26±0.25	ND	8.95±0.45	3.51±0.15	1.98±0.75
RSENc	332±1.10	4.95±0.15	1093±5.50	6.89±0.25	8.24±0.75	ND	13.1±0.50	0.55±0.05	2.88±0.35
NRSEAx	866±2.50	8.04±0.25	6332±2.25	228±1.54	53.6±0.15	9.28±0.05	89.1±0.15	9.43±0.25	8.47±0.55
NRSEAy	867±2.25	8.23±0.45	2928±2.25	221±1.66	53.7±0.15	8.35±0.05	87.2±0.15	8.30±0.25	10.4±0.55
NRSEAz	868±2.15	8.06±0.35	1356±2.25	221±1.50	53.6±0.25	7.25±0.07	83.2±0.15	5.27±0.25	11.0±0.55
DPR (2012)	28.0	0.80	0.50	200	36.0	35.0	29.0	85.0	100

The high levels of Cu, Cr and Pb in the non-remediated soils could be due to oxidation reactions in which the solubility of heavy metals increases with decreasing pH whereas in reducing conditions, the solubility of the heavy metals (Cu, Cd and Pb) is higher at high pH as a result of the formation of stable soluble organo-mineral complexes [20].

The concentration of Cr observed in this study were below those reported by [13] in his findings which had 4.71- 21.66 mg/kg while Pb, Cu and Cd were lower than those reported in this work. Soil pH is very important for most heavy metals since metal availability is relatively low when the pH is around 6 to 7. In the remediated soil, the availability of Se and

As were reduced in both dry and wet seasons with increasing soil pH because of the precipitation of these heavy metals as insoluble hydroxides, carbonates and organic complexes. At high pH, ion hydrolysis is favored, and the energy barriers that must be overcome when these ions approach the surface of soil particles decreases [21].

The results obtained shows that there is no significance difference in the metal concentration in remediated and non-remediated soils during dry and wet season with p- value (0.102) < 0.05.

4 Conclusion

The results obtained in this study revealed that, heavy metals concentration were higher at the non-remediated soil which recorded high pH values than the remediated soil. The remediation increases the pH of the soil which leads to the lower concentrations of some of the metals in both dry and wet seasons. However, the dry season recorded higher concentrations than the wet seasons. The results of the pH showed that the non-remediated soil were more acidic than the remediated soil in both seasons. Therefore, at low pH, the metals availability tends to increase as a results of oxidation reactions which leads to higher solubility and availability of metals in soil.

Compliance with ethical standards

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Disclosure of conflict of interest

The authors declare no conflict of interest.

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