Health Risk Assessment of Potentially Toxic Elements in Groundwater Samples within a Petroleum Depot Host Community in Ibadan, Oyo State, Nigeria

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The proximity of the Nigeria National Petroleum Cooperation (NNPC) Depot at Apata in Ibadan, Oyo State to human settlement in the Adebisi Layout area of Apata may lead to groundwater contamination which is the main source of potable water supply in this layout. Hence the need to assess the physicochemical status and concentrations of Potentially Toxic Elements (PTEs) in groundwater samples in this layout. The physicochemical status, as well as the concentrations of PTEs (Cd, Cr, Ni, Pb and Zn) in ten groundwater samples within the study area, were determined by standard methods and Atomic Absorption Spectrometry (AAS) respectively. The physicochemical parameters studied ranged as follows: pH 6.64 - 7.38, temperature 27.2 - 29.2 °C, Total Dissolved Solids (TDS) 0.99 - 1.60 mg L^{-1} and Dissolved Oxygen (DO) 1.05 - 1.65 mg L^{-1}. The concentrations of heavy metals in groundwater samples were in decreasing order of Cd > Ni > Zn > Cr, with concentrations ranging from 0.04 - 0.31 mg L^{-1} for Cd, 0.09 - 0.67 mg L^{-1} for Cr, ND - 0.04 mg L^{-1} for Zn and ND - 0.16 mg L^{-1} for Ni. The concentration of Pb was below the detection limit. Data from the non-carcinogenic risks showed that groundwater consumption may lead to adverse health effects and unacceptable risks to the human health of the local inhabitants, but data from the carcinogenic risks assessment indicate no cancer risk.

Keywords: Petroleum Contamination, Heavy Metals, Human Health Hazard, Groundwater Contamination.

1. Introduction

Safe drinking water is vital to maintain good health and a stronger nation (WHO, 2008), however, the constant increase in human population and rural-urban migration has placed a huge demand on this natural capital, creating scarcity. Additionally, improper water management practices can lead to the spread of water-borne diseases, which accounts for 6.3% of global deaths (WHO, 2008; Omole and

Ndambuki, 2014). About 9% of the world population lacks access to potable water while 2.4 billion cannot adequately access quality sanitation facilities despite the coordinated global effort to actualize the Millennium Development Goals (MDGs) target (UNICEF/WHO, 2015). Consequently, access to adequate potable water is highly imperative to promote public social welfare and development (Joseph, 2012).

Groundwater is one of the earth's most vital renewable and widely distributed resources, as well as an important source of water supply throughout the world. Groundwater accounts for about 98% of the world's fresh water and it is fairly well distributed across the globe (Sodde and Barrocu). Nearly two billion people (approximately one-third of the world's population) depend on groundwater supplies, withdrawing about 20% of global water (600-700 km^3) annually, much of which is from shallow aquifers. In Nigeria, urban and rural areas depend on groundwater as an indispensable source of adequate potable water. Moreover, the scarcity of municipal pipe-borne water in urban areas has encouraged the inhabitants to rely more on groundwater resources for daily use.

Nigeria is one of the major crude oil-producing countries in the world. It transports petroleum products through pipelines to several oil depots located all over the country from where it is carried by mobile tankers to end-users. The environment is contaminated with these products through accidental spills and leakages during the loading and offloading of tankers in the depots as well as the washing of oil storage tanks. All these discharges arise from several activities in the depot and significantly contaminate soil which ultimately pollutes both surface and groundwater through leaching or filtration, thereby constituting severe health and environmental hazards to humans and aquatic resources living in the area. The Nigerian National Petroleum Cooperation (NNPC) depot that supplies refined petroleum products to consumers is located in Apata, Ibadan, Oyo State. It is known that heavy metals such as Cd, Cr, Pb, Ni and Zn are normal constituents of petroleum (Fu et al., 2014) and several studies have linked elevated levels of toxic heavy metals in groundwater to petroleum contamination (Ogunlaja et al., 2019; Ogunlaja et al., 2018; Onojake and Frank, 2013). The proximity of the depot in this area to human settlements poses a potential threat to the environment and the immediate population primarily via the consumption from groundwater sources. Hence, the elemental investigation is vital.

One of the most critical environmental issues today is groundwater contamination, and between the vast diversity of contaminants affecting water resources, heavy metals receive particular concern considering their strong toxicity even at low concentrations (Hazrat et al., 2019; Tahir et al., 2019; Wendling, 2018; Masindi and Muedi, 2018). Previously, we reported on the elemental elevation and risk assessment of water sources within a petroleum refinery host community (Ogunlaja*et al*., 2018). In the present study, we used multivariate statistical analyses (principal component analysis (PCA) and Cluster analysis CA) to study the effect of the proximity of the NNPC Depot on human settlement. Additionally, the daily human exposure and the possible health risks associated with the toxic elements via consumption of water from the groundwater were also assessed through the ingestion pathway.

2. Materials and Methods

2.1. Description of the Study Area

The study area is located very close to the NNPC Apata Depot, Ibadan in Ido Local Government Area in Oyo State, Nigeria (Figure 1). Ido is the third largest Local Government Area in Ibadan, it is located between longitude 3 33'20? to3º51'11? and latitude 7º17'50? to 7º44'50? and with a population of 103,261 withan area of 986 km^2 (NPC, 2006). The catchment area covers longitude 3º47'13? to 3º49'41? and latitude 7º22'43? to 7º23'55?. Apata lies in the wet climatic region, and it records an average annual temperature of 26.8 °C with the highest monthly average temperature in March. August is the coldest month of the year with an average of 24.6 °C. The study area receives an average of 1131 mm of rainfall annually. The preliminary investigation in the study area showed that the inhabitants of this community depend on groundwater as the main source of drinking water.

2.2. Sample Collection and Preparation

Ten representative groundwater samples were collected following a simple randomization method from selected tube wells within the study area in Apata. Samples were transferred and stored in sealed 1 L plastic bottles and kept in a refrigerator until analyzed.

2.3. Chemical Analysis and Quality Control

The pH and Temperature were determined immediately using a pH meter fitted with a glass electrode and a portable calibrated mercury-in-glass thermometer. Total Hardness (TH) was determined using the complexometric method. Alkalinity was determined by titrimetry while conductivity, total dissolved solids (TDS) and dissolved oxygen (DO) were measured with a

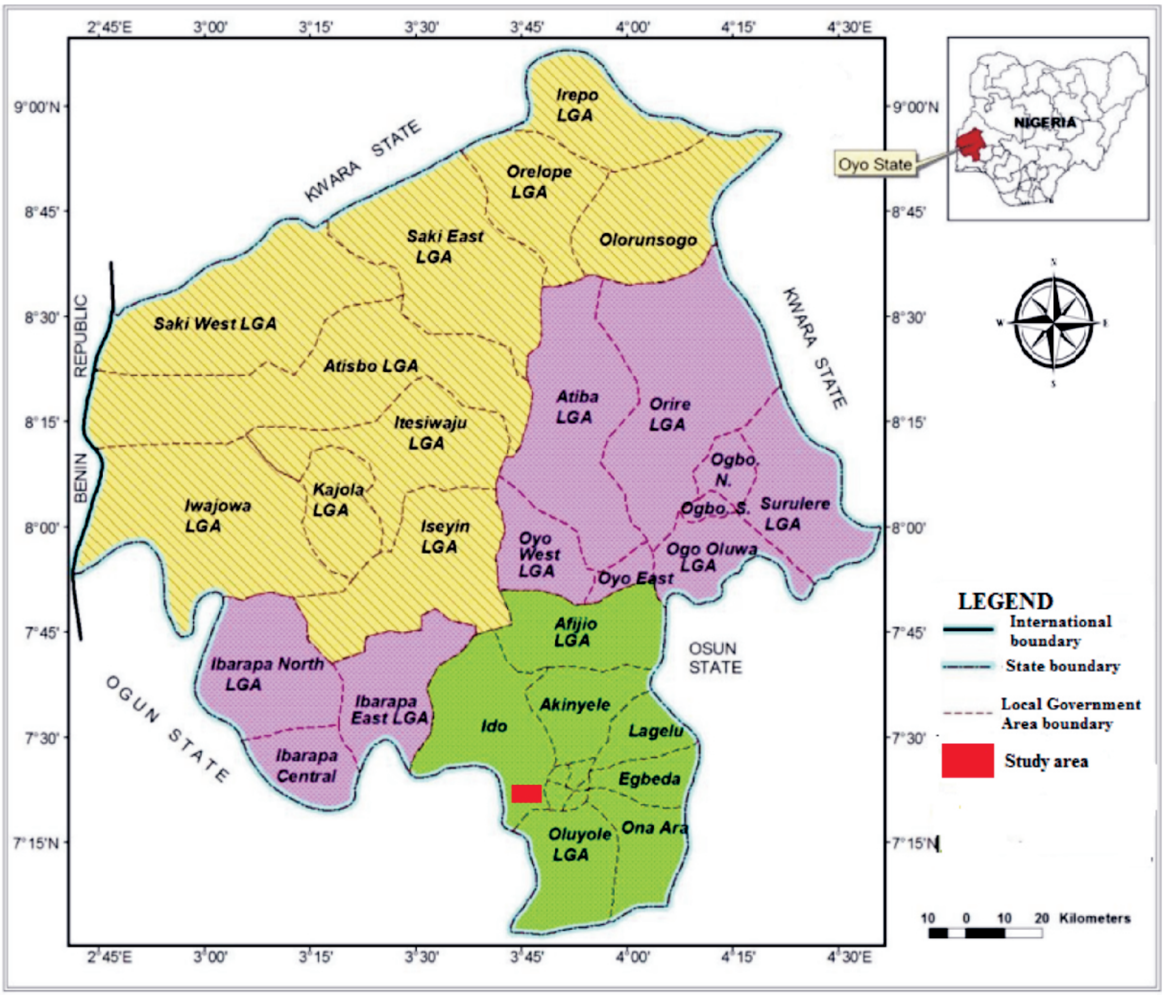


Figure.1: Map Showing the Study Area

membrane probe (Tutron WA-2015) supplied with an appropriate calibration solution. All the physicochemical results obtained were compared to permissible limits based on the World Health Organization (WHO) and Nigerian Standard for Drinking Water Quality (NSDWQ) standards (NSDWQ, 2007; WHO, 2011).

Heavy metals were determined by digesting a known volume of water samples with analytical grade HNO_3. The digested sample was filtered into a 25 ml standard flask, made up to the mark with deionized water and stored in a nitric acid pre-washed polyethylene bottle in the refrigerator before instrumental analysis. The water extracts were analyzed for metals (Cd, Cr, Pb, Zn and Ni) by the atomic absorption spectrometer (Schimazo model 2380). The detection limits were determined using instrumental parameters optimized for the individual element based on a 98% confidence level (3 standard deviations) and trace detection and increased sensitivity was used in all measurements to detect concentrations at the sub-ppb range for studied elements. Blank analyses were also carried out and the samples were analyzed in duplicates, and the average of the results was taken.

2.4. Reagents, Analytical Quality Assurance and Standards

Analytical reagent grade chemicals were used for samples and spectroscopic grades were used for standards. Elemental calibration standards were prepared from spectroscopic grade stock standard solutions of 1000 mg L^{-1}. For quality control, blanks were analyzed. Glassware and other equipment were cleaned with 6M HNO$_3$ and rinsed off with double distilled water to prevent contamination before usage. Deionized water was used throughout the experiments. All plastic containers were washed with double distilled water and then soaked overnight in 1M HNO$_3$.

2.5. Statistical Analysis

To investigate the possible sources of different metals from these aquifers, the concentrations of heavy metals in this study were analyzed using Pearson's correlation matrix. All statistical analyses were performed using the Statistical Package for the Social Sciences, (PASW version 24, IBM Corporation, Cornell, NY, USA).

2.6. Elemental Analysis

2.6.1 Health Risk Assessments

The chronic health hazard associated with the consumption of water from these groundwater sources was assessed. The daily human exposure assessment to heavy metals through the ingestion pathway was evaluated using the lifetime average daily dose (LADD), as adopted by USEPA (2005). In this study, the human exposure risk was estimated according to the modified equation from USEPA by Kavcar et al. (2009) and Belkhiria et al. (2017). The chronic risk was determined using chronic daily intake (CDI) and hazard quotient (HQ) index.

$CDI = (C \times DI) / (BW)$ .... (1)

Where CDI is the human exposure risk through ingestion pathway (mg/kg-day)$^{-1}$, C is the concentration of heavy metal in drinking water in mg L^{-1}, DI average daily intake rate (2.0 L/day-person)$^{-1}$ and BW is the body weight (15 kg and 72 kg for child and adult respectively).

The non-carcinogenic hazard was evaluated by the hazard quotient (HQ) by equation 2.

$HQ = CDI / RfD$ .... (2)

Where RfD is the oral reference dose (mg/kg-day)$^{-1}$ for individual heavy metal (Table 1) that humans can be exposed to, and for this study were obtained from USEPA. HQ is calculated for each heavy metal and the sum of HQ of all metals is used to determine the non-carcinogenic risk, hazard index (HI). If HQ < 1, it is considered safe for human health, 1? HQ $\leq$ 5 is low risk, 5 ? HQ $\leq$ 10 is medium risk and HQ ? 10 is regarded as high risk.

Table 1: The toxicity responses to heavy metals as the oral reference dose (RfD) and oral slope factor (SF)

Metals	Oral RfD[a] (mg/kg-day)$^{-1}$	Oral SF[b] (mg/kg-day)$^{-1}$
Cd	5.0×10^{-4}	3.8×10^{-1}
Cr	3.0×10^{-3}	5.0×10^{-1}
Pb	3.6×10^{-3}	9.0×10^{-3}
Zn	3.0×10^{-1}	ND
Ni	2.0×10^{-2}	1.7

[a]US EPA IRIS (2011), [b]USEPA (2015) and ND - not determined

Additionally, the carcinogenic risk was estimated. Carcinogenic risk characterization uses contaminant intake and toxicity index known as slope factor (SF) (Table 1) to compute the potential cancerous health risk. The cancer risk was estimated as the incremental probability of an individual developing cancer over a lifetime as a result of exposure to the possible carcinogen (Equation 3).

Target Carcinogenic Risk (TCR) = SF × CDI…………………………………….. (3)

Where the slope factor (SF) converts the chronic daily intake (CDI) to the incremental risk of individual developing cancer.

3. Results and Discussion of Findings

3.1. Physico-Chemical Parameters

The result of the physicochemical investigation of groundwater samples is summarised in Table 2. The pH values ranged from 6.64 to 7.38 and Figure 2A shows the distribution of pH across all sampling sites. These values indicated that the studied groundwater samples are mostly neutral and are within the acceptable limits of WHO and NDWQS guidelines. Water pH < 6.5 makes well water samples to be soft, and corrosive leading to elevated levels of toxic metals. Also, Figure 2B

Shows the distribution of DO within the studied groundwater samples which ranged from 1.05 to 1.65 mg L^{-1} and were all within acceptable WHO limits. The temperature also ranged from 27.2 to 29.2 °C and varied slightly from well A to J (Figure 2C).

The conductivity values ranged from 0.361 to 1.139µs cm^{-1} with significant variation from well A to J (Figure 2D). Similarly, the TDS values ranged from 0.99 to 1.60 mg L^{-1} with minimal to slightly significant variation in TDS values (Figure 2E). But, the TDS values from all the wells were within the acceptable limit of 1000 mg L^{-1}. Total Hardness values ranged from 25 to 151 mg L^{-1} and are significantly distributed within the studied groundwatersamples (Figure 2F). All the samples studied for TH were within acceptable limits, except Well C with the value of 154 mg L^{-1}. Furthermore, Alkalinity ranged from 0.07 to 0.17 mg L^{-1} and Figure 2G shows Alk. Distribution among the studied Well water samples. Alkalinity in water provides an idea of the natural salts present in water. Generally, there were no significant differences ($p > 0.05$) in the mean values for almost all the physicochemical parameters studied except for TH (Table 2). This observation suggests a common origin.

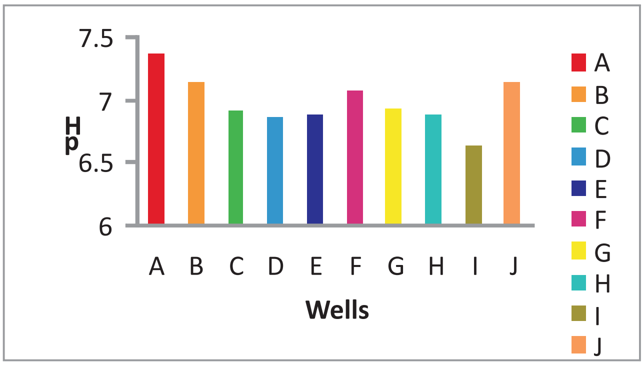
The values of the physicochemical parameters in this study were all within acceptable limits of WHO and NSDWQ drinking water standards respectively except in well C which ranged from 147 to 154 mg L^{-1} with a mean of 151 ± 5.0 mg L^{-1}. Hence, drinking from these wells may not pose a health risk to life.

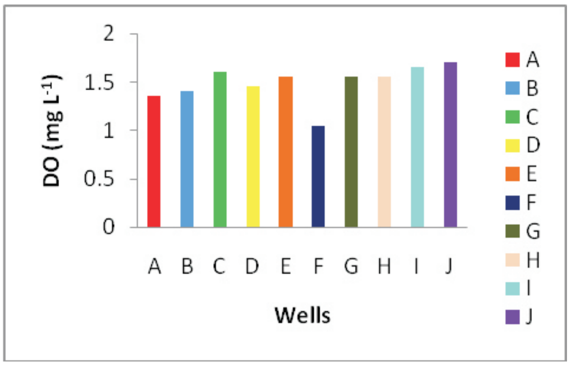
3.2. Elemental Analysis: The accuracy and precision of the analytical procedure were validated by the analysis of water CRM and experimental values compared well to the certified values ($p < 0.05$) (Table 3). Recoveries ranged from 99.8 to 101 %.

Table 3: Validation of the Analytical Method using Certified Reference Materials (CRM)

Certified Reference Materials	Elements	Measured (µg g^{-1})	Certified (µg g^{-1})	Recovery (%)
Water GBW08608	Cd	0.104 ± 0.002	0.104 ± 0.002	100.0
	Cr	0.509 ± 0.05	0.51 ± 0.01	99.8
	Cu	1.029 ± 0.01	1.03 ± 0.01	99.9
	Ni	0.516 ± 0.003	0.517 ± 0.006	99.8
	Pb	1.04 ± 0.08	1.03 ± 0.02	101.0
	Zn	5.14 ± 0.01	5.15 ± 0.05	99.8

*Values mean ± standard deviation, 95% confidence interval, n = 3





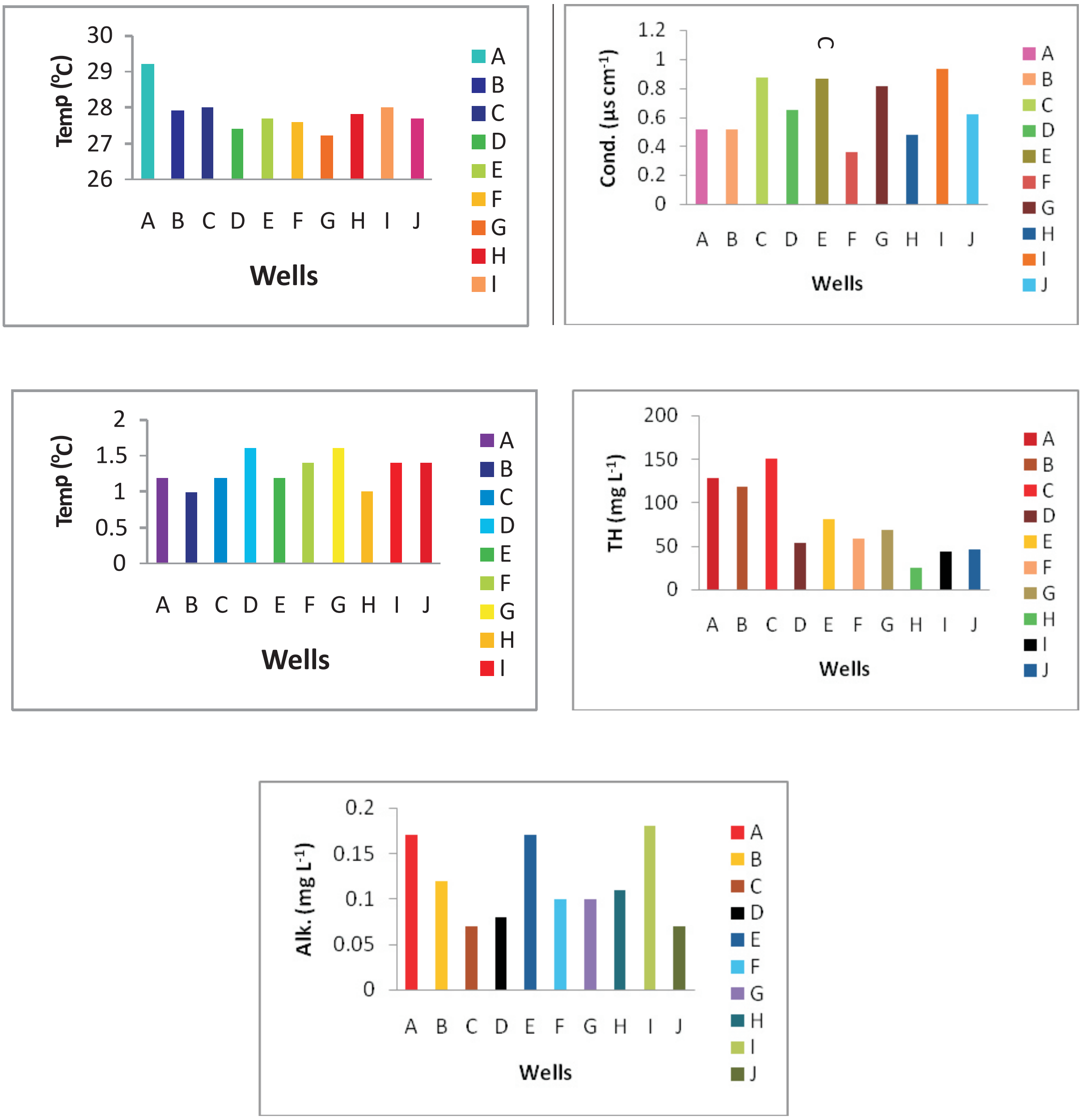


Figure 2: Distribution of Physicochemical Parameters in Studied Groundwater Samples

Well	**pH**	**DO (mg L^{-1})**	**Temp (^{0}C)**	**Cond. (µs cm^{-1})**	**TDS (mg L^{-1})**	**TH (mg L^{-1})**	**Alk. (mg L^{-1})**
A	7.38 ± 0.3^a	1.35 ± 0.1^a	29.2 ± 0.3^a	0.512 ± 0.02^a	1.20 ± 0.01^a	128 ± 2.1^a	0.17 ± 0.01^a
B	7.15 ± 0.7^a	1.4 ± 0.1^a	27.9 ± 1.6^a	0.515 ± 0.01^a	0.99 ± 0.02^a	118 ± 2.8^a	0.12 ± 0.01^a
C	6.92 ± 0.5^a	1.6 ± 0.1^a	28.0 ± 1.4^a	0.876 ± 0.01^{ab}	1.20 ± 0.01^a	$\mathbf{151} \pm 5.0^{ab}$	0.07 ± 0.01^{ab}
D	6.86 ± 0.4^a	1.45 ± 0.1^a	27.4 ± 2.3^a	0.647 ± 0.01^a	1.60 ± 0.02^a	54 ± 5.1^c	0.08 ± 0.02^{ab}
E	6.89 ± 0.8^a	1.55 ± 0.1^a	27.7 ± 1.9^a	1.139 ± 0.01^b	1.20 ± 0.01^a	81 ± 3.5^c	0.17 ± 0.01^a
F	7.08 ± 0.8^a	1.05 ± 0.1^{ab}	27.6 ± 1.9^a	0.361 ± 0.01^a	1.40 ± 0.02^a	59 ± 2.1^c	0.10 ± 0.01^a
G	6.93 ± 0.7^a	1.55 ± 0.1^a	27.2 ± 2.6^a	0.816 ± 0.01^b	1.60 ± 0.01^a	69 ± 7.1^c	0.10 ± 0.01^a
H	6.89 ± 0.5^a	1.55 ± 0.1^a	27.8 ± 1.7^a	0.477 ± 0.01^a	1.0 ± 0.01^a	25 ± 3.5^d	0.11 ± 0.01^a
I	6.64 ± 0.5^a	1.65 ± 0.1^a	28.0 ± 1.4^a	0.933 ± 0.01^{ab}	1.40 ± 0.02^a	44 ± 5.7^c	0.18 ± 0.01^a
J	7.14 ± 0.9^a	1.7 ± 0.1^a	27.7 ± 1.8^a	0.618 ± 0.01^a	1.40 ± 0.01^a	46 ± 2.8^c	0.07 ± 0.02^{ab}
NSDWQ	6.50-8.50	N/S	N/S	1000	500	N/S	N/S
WHO	6.50-8.50	6.0	N/S	25.0	500	100-150	? 120

Temp (Temperature), EC (Electrical Conductivity), TDS (Total dissolved solute), TH (Total Hardness), Alk. (Alkalinity), DO (Dissolved Oxygen). All values are presented as (mean ± SD) n = 3. N/S = Not specified, NSDWQ (Nigerian Standard for Drinking Water Quality, 2007), WHO (World Health Organization, 2011). Different superscript letters within columns indicate mean separations by Tukey's post-hoc tests at the 5% level.

Table 4 summarised the concentrations of Cd, Cr, Ni, Pb and Zn in groundwater samples. The concentrations of heavy metals in groundwater were found to be in decreasing order of Cd > Ni > Zn > Cr > Pb (Table 4). The mean elemental concentrations in groundwater samples for Zn and Ni ranged from ND – 0.04 mg L^{-1} and ND - 0.16 mg L^{-1} respectively, while Pb was below the detection limit for all groundwater samples (Table 4). The mean Cd concentration was higher than the WHO-recommended drinking water limit of 0.005 mg L^{-1} for all groundwatersamples except for E, F and I. This elemental elevation of concentration represents 70% of the total studied groundwater. Likewise, Ni concentration was higher than the WHO-recommended drinking water limit for 50% of the studied groundwater (Table 4). Drinking from these groundwater samples may lead to the accumulation of these heavy metals in human tissues over time. Accumulated Cd in the kidney can lead to kidney dysfunction (Baldwin and Marshall, 1999). Elevated levels of Ni has also been reported in animals living very close to a petroleum refinery, causing different kind of cancers (Wuana and Okieimen, 2011). The concentration of Pb was below the instrument detection limit hence it is omitted from Table 4.

Table 4 : Concentrations of Heavy Metals in Well Samples Compared with WHO Standards

	Ni	Zn	Cr	Cd
A	0.02 ± 0.01^{a}	ND	ND	**0.18**± 0.01^{a}
B	ND	0.01 ± 0.005 ^{a}	0.03 ± 0.001 ^{a}	**0.12**± 0.01 ^{a}
C	**0.08**± 0.02 ^{b}	0.01 ± 0.001 ^{a}	ND	**0.13**± 0.001^{a}
D	ND	ND	ND	**0.08**± 0.001 ^{a}
E	0.02 ± 0.01 ^{a}	0.04 ± 0.01 ^{a}	0.02 ± 0.001 ^{a}	ND
F	0.01 ± 0.006 ^{a}	ND	ND	ND
G	**0.11**± 0.02 ^{b}	0.04 ± 0.01 ^{a}	ND	**0.09**± 0.002 ^{a}
H	**0.15**± 0.05 ^{b}	ND	0.01 ± 0.005 ^{a}	**0.1**± 0.01 ^{a}
I	**0.16** ± 0.01^{b}	ND	ND	ND
J	**0.06** ± 0.02 $^{a\,b}$	ND	0.01 ± 0.005 ^{a}	**0.17**± 0.01 ^{a}
WHO	0.02	5.00	0.05	0.005

Values are in mg L^{-1} (Mean ± SD) and WHO (2017).

3.3. Human Health Risk Assessment

3.3.1. Hazard Quotient (HQ)

Contamination of groundwater is a serious environmental and health issue in many communities (rural and urban), and heavy metal is a significant contributor (Marcovecchio et al., 2007; Vodela et al., 1997). Hence, it is vital to assess the human health hazard as a result of possible heavy metal contamination associated with the consumption of water from these wells.

Table 5 summarised the non-carcinogenic risk (HQ) estimates for Cd, Cr, Ni, Pb and Zn for two age groups. Although the HQ (children and adult) for most of the heavy metals studied were ? 1, the HQ (child and adult) for Cd was ? 1, for most of the well water samples, except well E, F and I, indicating an unacceptable non-carcinogenic risk to human health. The non-carcinogenic hazard of Cd was higher in children than in adults and well A had the highest HQ value of Cd of 48.0 for adults and 10.0 for child respectively. Additionally, the HI indicated Cd as the dominant contaminant in the Well samples, contributing 97 % of the HI for children. Likewise, Cd and Ni were the dominant contaminants in the Well samples, contributing 46.9 and 52.4 % of the HI for adults respectively. Previously, Cd and Ni were also implicated in the water sources around a petroleum-contaminated area (Ogunlaja et al., 2019). This result indicates that the inhabitants might be exposed to some potential health risks through the intake of heavy metals in wells.However, the children are exposed to a higher non-carcinogenic risk than the adults according to the mean HI values due to the physiological differences. Moreover, the HI values were ? 1 across all water sources and age groups, suggesting an unacceptable risk of non-carcinogenic effects on the health of the local inhabitants (ECETOC, 2001).

Table 5: Non-carcinogenic Risk (Hazard Quotient, HQ) and Overall Toxic Risk (Hazard Index, HI)

Group	**HQ**	Ni	Zn	Cr	Cd	**HI**
Child	A	0.1	0	0	48.0	**48.1**
	B	0	0.004	1.3	32.0	**33.3**
	C	0.5	0.004	0	34.7	**35.2**
	D	0	0	0	21.3	**21.3**
	E	0.1	0.02	0.9	0	1.0
	F	0.1	0	0	0	0.1
	G	0.7	0.02	0	24.0	**24.8**
	H	1.0	0	0.4	26.7	**28.1**
	I	1.1	0	0	0	1.1
	J	0.4	0	0.4	45.3	**46.2**
Adult	A	0.03	0	0	10.0	**10.0**
	B	0	0.001	0.3	6.7	**7.0**
	C	0.1	0.001	0	7.2	**7.3**
	D	0	0	0	4.4	**4.4**
	E	0.03	0.004	0.185	0	0.2
	F	0.01	0	0	0	0.01
	G	0.2	0.004	0	5	**5.2**
	H	0.2	0	0.093	5.6	**5.9**
	I	53.3	0	0	0	**53.3**
	J	0.1	0	0.1	9.4	**9.6**

The TCR for Ni, Cr and Cd is summarised in Table 6 and ranged from 0 to 18.2 for children and 0 to 90 for adults. The elemental TCR values were in the decreasing order of Cd ? Ni ? Cr and Ni ? Cd ? Cr for child and adult respectively. Generally, the TCR values of exposure of Ni, Cr and Cd to both adults and children were found to exceed the USEPA recommended safe limit for cancer risk (1×10^{-4}), indicating a chance of cancer risks (USEPA, 2011 and 2012). The cumulative risk ($\sum$ TCR) of exposures of Ni, Cr and Cd indicated that adults have higher chances of getting the risk of cancer than children. For children, Cd contributes 91.1 % to the total carcinogenic risks than Ni (7.2 %) and Cr (1.6 %) while for adults, Ni contributes 83.1 % to the total carcinogenic risks than Cd (16.6 %) and Cr (0.3 %).

Table 6: Target Carcinogenic Risk (TCR) of the Elements in Well Water Samples

Age group	**Cancer risk (TCR)**				**$\sum$TCR**
		Ni	Cr	Cd	
Child	A	0.23	0	18.2	18.5
	B	0	0.67	12.2	12.8
	C	0.91	0	13.2	14.1
	D	0	0	8.11	8.11
	E	0.23	0.44	0	0.67
	F	0.11	0	0	0.11
	G	1.25	0	9.12	10.4
	H	1.7	0.22	10.1	12.1
	I	1.81	0	0	1.81
	J	0.68	0.22	17.2	18.1
Adult	A	0.05	0	3.8	3.85
	B	0	0.14	2.53	2.67
	C	0.19	0	2.74	2.93
	D	0	0	1.69	1.69
	E	0.05	0.09	0	0.14
	F	0.02	0	0	0.02
	G	0.26	0	1.9	2.16
	H	0.35	0.05	2.11	2.51
	I	90.7	0	0	90.7
	J	0.14	0.05	3.59	3.78

A-J-Well water

Conclusion

The concentrations of studied heavy metals in water samples from Apata were found to be in decreasing order of Cd > Ni > Zn > Cr > Pb, respectively. This study showed that there is a significantly elevated level of some heavy metals such as Cd and Ni, which are normal constituents of petroleum. Thus, a potential pollution risk may exist, which might contribute to the heavy metal loading of groundwater in the study area. In addition, Health risk assessment data of groundwater consumption from this study indicated an unacceptable non-carcinogenic risk to the human health of the local inhabitants.

Recommendation

The results from this study may be used to educate petroleum Depot host communities, on groundwater consumption safety, in order to prevent adverse human health risks from potentially contaminated groundwater.

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