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Metals Contamination of Groundwater Resources of Enugu North District, SouthEast Nigeria

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KEYWORDS

Contamination;
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Seasonal variation

ABSTRACT: Ground water samples were collected from boreholes in Enugu Northern District, South-east Nigeria and analyzed for some metals (Cd, Cu, Fe, Pb, K, Na and Zn) contents. The metals were determined at monthly intervals for three months in dry season and then repeated in rainy season using standard methods. The range of values obtained during dry season include: Na (0.0–21.28 mg/L), K (0.0–12.03 mg/L), Cu (0.0–23.01 mg/L), Fe (0.0–11.03 mg/L), and Zn (0.01–3.00 mg/L). During rainy season, the metal contents were of the following ranges: Na (0.0-21.28 mg/L), K (1.32-15.11 mg/L), Cu (0.03-28.37 mg/L), Fe (0.0-12.29 mg/L), and Zn (0.02-3.61 mg/L). Cd and Pb were not detected in all the samples studied. The mean values of the parameters were compared with recommended standard guideline values and some were above the guideline values for drinking water quality. Seasonal variations of the parameters in the samples were observed with rainy season values higher. A two-way analysis of variance (ANOVA) using SPSS Windows Version 20 analysis showed statistically significant variations between metal contents in dry season and rainy season. Metal evaluation indices (MEI) indicated spatial and seasonal variations. Degree of metallic contamination computed showed that Cu and Fe were found to be in very high degree in both seasons.

INTRODUCTION

Groundwater form borehole is the predominant source of water for drinking and domestic use in Enugu North District, South-east Nigeria. Majority of the inhabitants in communities within this district daily consume untreated groundwater by exposing them to contaminants due to unsafe water. An estimated 65 million Nigerians do not have access to clean, reliable potable water [1]The high overall mortality rate due to water-related avoidable toxins and water-borne diseases currently being recorded globally has evoked a projection of about 135 million related human deaths by the year 2020 [2] in spite of the achievement of the Millennium Development Goals (MDGs) announced by the United Nations in 2000 [3]. There have been reported high infant mortality rate due to the consumption of polluted water [4, 5]. The collaborative efforts by developing countries and industrialized nations with organizations such as United Nations Environmental Programme (UNEP), World Health Organization (WHO), etc, are mainly in areas of

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monitoring and controlling the causes of water pollution and developing strategies for safe water supply and use [6 - 8].

Groundwater aquifers are prone to pollutants of agricultural, domestic and industrial origin; especially in areas of shallow water beds and high amounts of rainfall [9.10]. Studies have reported water-based toxic levels of some metals such as cadmium, chromium, manganese and nickel in blood samples of inhabitants of Enugu, Nigeria [10 - 12]. Some heavy metals such as arsenic, cadmium and lead are known to be biologically non-essential nutrients which may be found in water. Other essential metals such as copper, potassium, sodium and zinc become toxic at high concentrations in the body.

Clinical diagnosis of metal toxicity in humans is usually a difficult task as a result to the nature of the symptoms expressed thereby making baseline data of potential sources of metal poisoning such as contaminated groundwater essential to serve as first-hand diagnostic information when needed [13]. Presently, no single assessment of metals in groundwater has covered Enugu North District. However, a recent study showed seasonal variations in Biochemical Oxygen Demand (BOD), Chemical Oxygen Demand (COD) and elevated temperatures in the aforementioned area [14]. The aims of this work were to assess the concentrations and seasonal variations of some metals in ground water resources in the study area with the view to establishing a comprehensive base line data for such metals in the area.

MATERIALS AND METHODS

Study Area

The study was conducted in Enugu North District, an area with an estimated population of 1,377,001 covering Igbo Etiti, Igboeze North,IgboezeSouth, Isi-uzo, Nsukka, Udenu and Uzo-Uwani Local Government Areas [15]. A ten-day reconnaissance survey of the twelve towns and communities within the study area was conducted to identify sites for sample collection and marked with Geographical Positioning System (GPS Garmin 724). Table 1 shows the designations of sample points, their GPS locations and surrounding features.

Table 1. Study Area (sample location and surrounding features)

Sample Designation	LGAs	Town/GPS Coordinates	Surrounding features
BH1– 10	Igbo-Eze North	Enugu-Ezike (N: 6 ^{0} 41'24" E: 7 ^{0} 06' 53")	Farmlands, residential houses, automobile roads.
BH11- 20	Nsukka	Nsukka(N: 6 ^{0} 51'24" E: 7 ^{0} 23' 01")	Farmlands at pumping sites.
BH21-30	Udenu	Obolo (N: 6 ^{0} 34'08" E: 6 ^{0} 27' 54")	Residential houses, market, busy vehicular activities/roads.
BH31-40	Igbo-Eze South	Ovoko (N: 06 ^{0} 41'33" E: 007 ^{0} 11' 04")	Residential houses, farmlands and busy vehicular activities/roads.
BH41-50	Igbo-Etiti	Ogbede (N: 5 ^{0} 23'46" E: 6 ^{0} 30' 12")	Market, surrounding farmlands, residential houses, busy vehicular activities/roads.
BH51-60	Nsukka	Opi(N: 06 ^{0} 43'36" E: 007 ^{0} 13' 08")	Petrol station, cement block factory, farmlands, automobile and metal bending workshops.
BH61-70	Isi-Uzo	EhaAmufu(N: 06 ^{0} 42'35" E: 007 ^{0} 13' 05")	Refuse dump site; nearby farmlands.
BH71-80	Nsukka	Eha-Alumona (N: 06 ^{0} 43'35" E: 007 ^{0} 13' 07")	Surrounding farmlands.
BH81-90	Uzo-Uwani	Adani(N: 06 ^{0} 43'30" E: 007 ^{0} 13' 00")	Residential houses and distant farmlands
BH91 100	Nsukka	Ede-Obala (N: 6 ^{0} 43'35" E: 007 ^{0}13' 09")	Aluminium sheets warehouse, automobile repair shops, district hospital, block-moulding industries, hotel, metal-welding workshop, etc.

Table 1. Continued

BH101-110	Nsukka	Ibagwa-Agu (N: 06 ^{0} 41'34" E: 007 ^{0} 11' 06")	Farmland and residential houses.
BH111-120	Nsukka	Okpuje (N: 06 ^{0} 41'30" E: 007 ^{0} 11' 02")	Ongoing building sites block factories, metal welding workshops and farmland.

BH = Borehole Location, LGAs = Local Government Areas

Sampling

The samples were collected from one hundred and twenty different sample points in the seven Local Government Areas in Enugu North District. Samples were collected in the months of December 2014 to February 2015 for Dry season and May 2015 to July 2015 for Rainy season samples. Samples were collected twice monthly (every first and last week of the month). This gave a total of 12 samples per borehole.

Groundwater samples from the boreholes were collected in clean sterilized 250 mL Nalgene plastic bottles with tightly fitting. At the collection points, the sample containers were meticulously rinsed with the borehole water before filling them with the sample. Before collection of the water samples, the water taps were purged by allowing the water to run for 10 minutes.

The collected water samples were preserved by adding 5 mL concentrated HNO_3 (65%, Merck) to prevent metals from adhering to the walls of the containers. The bottles were corked, labeled and refrigerated at 4°C in the laboratory prior to analysis. Filtration of samples was done using 0.45 mm millipore membrane paper placed in an all glass millipore filtering system. The membrane filters were washed with 1% (v/v) HNO_3 followed by rinsing in high purity water prior to filtration.

Quality Assurance

Recovery Analysis was performed using four randomly selected samples to determine instruments working conditions and method accuracy. Water samples were spiked with known amount of the pure metals salts, Calibrations were carried out using 1000mg/L stock solutions of the respective salts supplied by Merck, Germany. The stock solutions were serially diluted to achieve required concentrations for calibrations. R^2 values ranging from 92 to 98% were recorded for the metals.

Instrumentations

Sodium and potassium were determined using APHA 3500-K B and APHA 3500-Na B standard flame photometric methods respectively [16] with a Sherwood Flame Photometer Model 360 (Sherwood Scientific Ltd, Cambridge. Heavy metals were determined using Graphite Furnace Atomic Absorption Spectroscopy (GFAAS) in accordance with standard procedure [17] with a Perkin Elmer PinAAcle 900T Atomic Absorption Spectrophotometer (PerkinElmer Inc. USA).

Data was analysed using Statistical Package for Social Scientists (SPSS windows version 20) and Analysis of Variance (ANOVA), two-factor analysis and Dunnett's Post Hoc test were determined.

RESULTS

Table 2 shows the concentration (mean ± standard deviation and ranges) of metals in boreholes water samples from the twelve sampling areas during rainy season and dry season. The range of percent recoveries for the metals recorded were Cd =98.85 – 99.96; Cu = 89.49 – 99.97; Fe = 99.22 – 99.96; Pb = 99.86 – 99.98; Zn = 91.68 – 99. 81; K = 96.00 – 99.39 and Na = 99.00 – 99.37. The high percentages of metals recovered in the selected water samples studied attested to the accuracy of the methods and instruments used in the analysis.

Table 2. Results (Mean ± SD) of the Metals Concentrations (mg/L) for Dry Season and Rainy Season.

BHSZ	Cd	Pb	Fe	Cu	Zn	Na	K
01							
DS	bdl	bdl	0.1±0.2	22.3±0.1	2.6±0.2	10.0±0.0	6.0±0.0
RS	bdl	bdl	0.1±0.0	24.2±0.1	2.8±0.1	12.2±0.2	10.1±0.0
02							
DS	bdl	bdl	0.1±0.1	bdl	0.02±0.0	3.3±2.8	1.9±0.0
RS	bdl	bdl	0.1±0.0	0.0±0.0	0.03±0.0	3.4±0.2	2.1±0.0
03							
DS	bdl	bdl	0.3±0.0	22.5±0.8	0.8±0.1	12.3±0.0	7.1±0.0
RS	bdl	bdl	1.3±0.0	27.5±1.1	2.1±0.2	13.3±0.1	11.1±0.0
04							
DS	bdl	bdl	0.7±0	1.6±0.2	0.01±0.0	2.0±0.0	8.4±0.0
RS	bdl	bdl	1.1±0.0	2.6±0.2	0.08±0.1	2.1±0.0	11.0±0.1
05							
DS	bdl	bdl	bdl	0.01±0.0	0.01±0.0	2.0±0.0	8.4±0.0
RS	bdl	bdl	bdl	0.1±0.0	0.02±0.0	2.1±0.0	11.0±0.1
06							
DS	Bdl	bdl	0.5±0.1	19.2±0.8	2.4±0.2	3.1±0.1	bdl
RS	bdl	bdl	1.6±0.1	21.5± 0.3	3.4±0.2	4.1±0.1	12.0±0.0
07							
DS	bdl	bdl	10.5±0.4	16.6±0.2	2.1±0.0	22.4±0.0	7.7±0.0
RS	bdl	bdl	12.3±0.1	24.6±.6	0.1±0.0	21.2±0.0	15.1±0.0
08							
DS	bdl	bdl	0.1±0.1	10.7±0.01	1.7±0.2	13.9±0.1	17.2±0.0
RS	bdl	bdl	0.2±0.1	18.3±0.7	2.4±0.1	14.2±0.1	15.2±0.0
09							
DS	bdl	bdl	0.1±0.0	0.3±0.0	0.1±0.0	13.9±0.0	6.5±0.0
RS	bdl	bdl	0.1±0.0	0.4±0.1	0.1±0.0	14.1±0.1	11.2±0.0
10							
DS	bdl	bdl	0.1±0.0	0.1±0.1	0.2±0.0	18.6±0.1	9.2±0.0
RS	bdl	bdl	0.1±0.0	0.030.0	0.2±0.0	17.1±0.0	10.0±0.0
11							
DS	bdl	bdl	0.8±0.6	12.5±0.7	3.0±0.0	13.9±0.0	6.5±0.0
RS	bdl	bdl	0.1±0.0	13.0±0.7	2.9±0.1	14.1±0.1	11.2±0.0
12							
DS	bdl	bdl	1.6±0.5	0.1±0.0	2.9±0.0	18.6±0.2	8.1±0.0
RS	bdl	bdl	0.1±0.0	0.1±0.0	2.8±0.0	19.0±0.1	14.0±0.0
WHO permissibl e limi	0.003	0.01	*0.300	2	*30.	50	10

BHSZ = Bore hole Sampling Zone comprising of 10 boreholes. DS = Dry season, RS = Rainy Season, Bdl = below detection level
* Values are given by Nigerian Standard for Drinking Water Quality (NSDWQ).

Table 3 shows the degree of metallic contamination of the water samples in dry season (DS) and rainy season (RS) of the year 2015.

Table 3. Degree of Metallic Contamination of the studied Groundwater

Metals	**Cd**		**Pb**		**Cu**		**Fe**		**Zn**		**Na**		**K**	
Season	DS	RS	DS	RS	DS	RS	DS	RS	DS	RS	DS	RS	DS	RS
C^d	-1	-1	-1	-1	94	121	12	45	-2	-6	-11	-11	-4	0.2

C^d = Degree of Contamination, DS = Dry Season, RS = Rainy Season.

DIA

Cadmium was not detected in all the water samples during dry season and rainy season as shown in Table 2. Many studies of borehole water in some parts of south-east Nigeria and Enugu state in particular showed similar results of the absence of Cd (or below detection limit) but was detected in some water samples from other sources [18 - 23].. As shown in Table 2, Cu ranged between bdl - 23.01 mg/L during the dry season with a mean concentration of 8.9±0.3 mg/L which increased to 11.1±1.2 mg/L during rainy season. This may result from a change in corrosive water characteristics that lead to increase in copper solubility. Such characteristics include a decrease in pH below 6.5 and an increase in acidity which may lead to corrosion of copper-containing distribution piping systems. The dry season and rainy season mean content of Cu in the water samples were above the recommended guideline value of 1.00 mg/L.[25].The Cu content was also above those obtained in borehole water samples from some parts of the South-east area of Nigeria from previous studies. [16, 18, 19, 24 and 25].

The levels of Fe in the water samples during dry season and rainy season indicated a slight rise in iron content during rainy season and these were above the recommended guideline value of 0.30 mg/L.[25]. Fe content in the area studied has no specific spatial pattern. Anthropogenic contamination sources as well as increased solubility of Fe due to low pH during rainy season may be implied. Similar studies [16, 23] recorded higher iron concentrations in shallow hand dug wells, lake/ponds and rivers in some rural regions of South-east Nigeria and some deep aquifers in Nsukka south-east . However, Engwa et al., [18] recorded “not detected” in all the water sources from Enugu metropolis studied for Fe content (including water from borehole source) while Okoye and Adiele [19] recorded Fe concentrations in borehole water sample in an area in Imo state Nigeria lower than the recommended guideline value. Fe in borehole water was also found to be at lower concentrations in some borehole water samples from Abia state, south-east Nigeria and Akwa-Ibom state, South-South Nigeria.

Pb was below detection level in all the water samples during dry season and rainy season. Such result was obtained in all but one borehole water sample collected from various locations within Ebonyi, a neighboring state to Enugu state, south-east Nigeria [24]. However, studies of some borehole water samples from Enugu Urban and water samples from some shallow hand-dug wells, rivers and lakes/ponds clusters in some rural regions of south-east Nigeria had Pb above the maximum permissible level [16, 18].

Zn levels in all the boreholes water samples slightly increased maximally from 1.3±1.2 mg/L recorded in dry season to 1.4±0.4 mg/L in rainy season. These mean Zn values in both seasons were below the maximum permissible level of 3.00 mg/L. set by WHO [25]. Some studies also recorded low Zn content in borehole water from Enugu urban [16] and Imo state [19] of Eastern Nigeria. However, study of some water samples collected from boreholes and hand-dug wells in Benin City, Nigeria found Zn above the recommended guideline value [26].

The mean values concentration of Na were below the recommended maximum permissible level of 200 mg/L Na in drinking water set by SON [26] and the European Union (EU) [27]. Similar results were recorded in independent studies of the quality of groundwater resource collected from Imo state [19].

The mean concentration of K increased from 6.4±30.7 mg/L observed in dry season to 10.2±0.2 mg/L in rainy season. Seasonal variability of K content in the groundwater may result from leaching of dissolved potassium from fertilizers into groundwater during the rainy (crop planting) season. The rainy season mean value is slightly above the standard guideline value of 10.0 mg/L recommended by the EU [27]. The content of potassium in some hand-dug wells and springs of discharge farmland settlement in Nsukka south-east were in the range of 0.0 - 10.0 mg/L [23] while another report showed a range of 1.00 – 12.00 mg/L potassium in borehole water from Imo state both in south-east Nigeria [19].

STATISTICAL ANALYSIS

The ANOVA of the metal contents during dry season showed significant difference between the boreholes water samples (F (6, 77) = 12.39, $p < .05$). A Dunett's T3 post-hoc test of the dry season metal results revealed a significant difference due to Cu ($p< .05$), potassium ($p< .05$) and sodium ($p< .05$) mean contents while each of the mean concentrations of iron and zinc metals were relatively the same in the water samples: Fe ($p< .93$), Zn ($p< .06$). There was also significant difference in the water samples during rainy season due to their metals contents as shown by the ANOVA result (F(6, 77) = 12.91, $p < .05$). A Dunett's T3 post-hoc test of multiple comparison showed that the mean contents of the heavy metals: Cu ($p < .10$), Fe, ($p < .91$), Zn ($p < .08$), Cd ($p < .10$) and Pb ($p < .10$) in the 12 water samples were relatively the same but the samples significantly varied due to the mean contents of potassium ($p < .05$) and sodium ($p < .05$). The concentration of metals in the samples during dry season is significantly different from the rainy season metals results as revealed by the ANOVA (F (13, 154) = 11.81, $p < .05$). Dunett's T3 showed Cu (p = 1.00), Fe (p = .100), Zn (p = .100), potassium (p = .79), Sodium (p = .100).

Estimation of Metal Evaluation Indices (MEI) and Degree of Metallic Contamination of the water Samples

Metal Evaluation Indices

The Metal evaluation Index (MEI) of each metal in the 12 clusters borehole water samples was calculated using the formula [28] below:

$$MEI = \sum_{i=1}^{n} \frac{Mc}{Mmax}$$

where: Mc = Analyzed concentration of the respective metal;

and$Mmax$= Maximum permissible concentration of the respective metal.

MEI is an indication of the overall quality of the water in relation to the metals estimated This study assumed that the MEI value of a pure sample is unit (1.0); Then, MEI value of a metal above the one an indication of pollution. Thus groundwater metal activities due to Cu (1160.11 in dry season and 1320.65 in rainy season); Fe (490.75 in dry season and 560.46 in rainy season) and K (120.23 in rainy season) were higher than 120. When seasonal values of MEI were expressed as quotients to indicate the extent of seasonal variation due to the water quality parameter from a specific source or area studied,, the ranking of seasonal variation due to the metals in the samples from the area studied is as follows: Zn (1.07) < Fe (1.13) < Na (1.18) < Cu (1.25) < K (1.59). Zn showed the lowest variation gap and potassium had the highest variation. This may be due to the higher use of potassium-rich fertilizers which seep into leaky (cracked) groundwater aquifers or via water exchange points during the rainy season than dry season. Variation gaps due to copper may be as a result of increase in cuprosolvency of the water caused by drop in pH values.

Degree of Contamination

The degree of metallic contamination, $\mathbf{C^d}$ of the borehole water in the study area was estimated using the equation [29]:

$$C^d = \sum_{i=1}^{n} \frac{C_a}{C_{max}} - 1$$

Where: $\boldsymbol{C_a}$is the analytical concentration value of the respective metal.

C_{max} is the maximum permissible concentration of the respective metal.

The degree of contamination of water is categorized into ‘low’ ($\mathbf{C^d < 1}$), ‘medium ($\mathbf{C^d}$ ranges from 1 to 3) and ‘high’ ($\mathbf{C^d > 3}$) [30]. Cd and Pb were undetected in the samples and recorded low degree of contamination. Contamination due to K, Na, and Zn were low in both seasons but very high for Cu and Fe. The degree of metal contamination in the water during dry season trended as follows: Na < K < Zn < Cu < Fe and in rainy season: Na < Zn < K < Cu < Fe.

CONCLUSIONS

Metallic activities in the groundwater samples were mostly due to Copper (Cu), iron (Fe), sodium (Na), potassium (K) and zinc (Zn). The metallic contamination of the domestic groundwater in Enugu north senatorial district may be due to inputs from municipal waste water percolation, industrial as well as agricultural nutrients sources. Seasonal as well as spatial variations of the boreholes water in terms of the concentration of each metal were observed as revealed by the Analysis of variance, post hoc tests and the metal evaluation indices. The mean concentrations of Cu and Fe were above recommended guideline values and showed very high degree of contamination in both dry and rainy seasons.

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