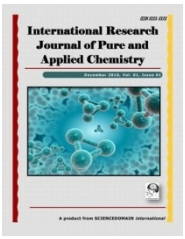

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Explanatory Interaction Profile of Cd, Pb and Zn on the Relative Abundance of Fe as Response Variable in Drinking Water Quality Assessment

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Authors' contributions

This work was carried out in collaboration between all authors. Author UM designed the study, managed sample preparation and instrumental analysis. Author MW managed sample collection and transportation to the laboratory. Author SSH performed the statistical analysis, managed the literature searches, wrote the protocol and prepared the manuscript. All authors read and approved the final manuscript.

Research Article

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ABSTRACT

Aims: The present study determined the quality status of drinking water in some locations of Northeastern Nigeria and attempted to present an explanatory interaction profile of Cd, Pb and Zn on the bases of the relative abundance of Fe in the drinking water samples.
Study Design: The study was designed to follow a two step procedure viz. quality assessment of drinking water samples and explanatory interaction profile of Cd, Pb and Zn on the relative abundance of Fe using multiple linear regression analysis on quality data obtained.
Place and Duration of Study: National Agency for Food and Drug Administration and Control (NAFDAC) zonal laboratory, Maiduguri. April 2007 to June 2008.
Methodology: Random grab water sampling technique was adopted. The determination of Fe Cd, Pb and Zn in drinking water samples were performed using the standard methods of Flame Atomic Absorption Spectrometry. Analyse-it (version 2.26) statistical software for Microsoft Excel was used to perform multiple linear regression (MLR)

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analysis for the explanatory interaction profile study.
Results: Result of drinking water quality status show that Fe was highest in all locations as anticipated and ranged between 1.317 ± 0.0192 mg/L and 0.14 ± 0.003 mg/L. Zn ranged between 0.015 ± 0.008 mg/L and 0.007 ± 0.004 mg/L. Cd concentration ranged from 0.004 ± 0.002 mg/L to 0.059 ± 0.001 mg/L while the mean Pb concentration ranged between 0.074 ± 0.021 mg/L and 0.001 ± 0.001 mg/L. Spectacularly Pb was not detected in all water samples from one of the three locations. MLR analysis revealed Pb as a key agent of reducing Fe concentration in the water systems and its impact tend to be more rigorous when the concentrations of Cd and Zn are comparatively higher than it.
Conclusion: The result of this study indicated the need for Contaminant Control Programmes, specifically for Cd and Pb in ground water. The findings of the MLR analysis show that Fe varied diversely due to difference in the aquifer location concentrations of all three explanatory variables. Fe was found to be potentially lowered by the shared presence of all three explanatory variables, which was greatly influenced by the presence of Pb.

Keywords: Water quality; heavy metals; multiple linear regression; modeling.

1. INTRODUCTION

Iron (Fe) accounts for about 5% of the earth's crust, making it the second most abundant metal [1-3], hence the reason why it is found in most environmental matrices.

Cadmium (Cd) naturally occurs with Zinc (Zn) and Lead (Pb) in especially sulfide ores. Cd and Zn are chemically similar with an oxidation state of +2 [3]. The interaction profile, though not in isolation, for these metals in drinking water quality assessment have been reported [4-7], which in most cases indicate the presence of Fe in association with Cd, Pb and Zn. But none of these, as well as other related studies have considered closely the influence of interaction variations of Cd, Pb and Zn on the relative abundance of Fe in drinking water quality assessment.

The present study therefore attempts to present an explanatory interaction profile of Cd, Pb and Zn on the bases of the relative abundance of Fe in drinking water quality status. In other words, this study endeavours to answer the question: How does the relative abundance of Fe vary as the interaction profile of Cd, Pb and Zn varied in water quality status? The findings of this study is aimed at providing a functional modelling tool for estimation and predicting contamination levels of these metals in drinking water quality assessment, from diverse sources and locations.

Assessing drinking water quality is a sustainability management practice, in addition to elucidating health implications. This has received further relevance through modelling [8-9], especially in tackling aspects and variety of processes that lead to the rapid degradation of drinking water resources.

1.1 Background: Levels in Water and Health Implications of Fe, Cd, Pb and Zn

A general median value of 0.7 mg/L Fe concentration has been reported in rivers, while in anaerobic groundwater where iron is in the form of Fe (II), concentrations usually ranges between 0.5 and 10 mg/L, but concentrations up to 50 mg/L have been recorded [10]. Fe

concentrations in drinking-water are usually less than 0.3 mg/L, which may vary upwards due to applications of Fe-salts in coagulating agents, cast iron, steel, and galvanized iron pipes used for water supply. However, Fe is an essential trace element in living organisms. In humans, the major fraction is present as haemoglobin, myoglobin, and haem-containing enzymes. The other substantial fraction is stored in the body as ferritin and haemosiderin, primarily in the spleen, liver, bone marrow, and striates muscle [10-11]. On one hand, the average lethal dose of Fe is 200–250 mg/kg of body weight, but fatality has occurred subsequent to the drinking doses as low as 40 mg/kg of body weight. On the other hand, adults have frequently taken Fe supplements for extensive duration of time without harmful effects [12], while an intake of 0.4–1 mg/kg of body weight per day is implausible to be associated with adverse effects in healthy persons [10,13].

The concentrations of Cd in uncontaminated natural waters are typically less than 1 µg/L [14]. Data of Cd concentration collected form 110 sampling stations around the world was reported to have a median concentration of dissolved Cd to be less than 1 µg/L, while the maximum value 100 µg/L was recorded in Rio Rimao, Peru [15]. Drinking water contamination by Cd may occur as a result of the presence of Cd as an impurity in the zinc of galvanized pipes or cadmium-containing solders in fittings, water heaters, water coolers and taps [3]. The concentration of Cd has been reported to increase with age, and both kidney and liver act as Cd repositories; kidney stores up to 50–85% and liver, 30–60%. The estimated lethal oral dose for humans is 350–3500 mg, but a dose of 3 mg has no acute effects on adults [16]. Cd is considered as category I carcinogen [17]. Its concentration has been linked with lung [18], pancreases [19] and kidney cancer [20]. The Provisional Tolerable Weekly Intake (PTWI), a reference value for exposure and related health risk for Cd, is 7 µg per kg body weight, applicable both to adults, and to infants and children [21].

Pb occurs in tap water to a degree, consequent upon its dissolution from natural sources, but mainly from home plumbing systems in which pipes, solder, fittings or supply connections to homes contain Pb. Even polyvinyl chloride (PVC) pipes also have Pb compounds that can be leached into tap water supply system. Exposure to Pb through water is usually low compared to air or food. Results of analyses of both surface and ground water suggest that Pb concentration is rather low [22]. The WHO [23] maximum permissible contaminant levels for Pb in drinking water is 0.01 mg/L. Pb is a potentially toxic substance with no known physiological function. Pb toxicity affects the haematologic, renal and neurologic systems and there is no evidence for a threshold below which Pb has no adverse effects, especially in children health [24-25].

Zn concentration in natural surface waters is usually less than 10µg/L while in ground waters it ranges between 10 and 40µg/L [26-27]. In tap water, Zn concentration is comparably higher due to leaching from piping and fittings [28]. The contribution of drinking water to Zn intake is usually small except high concentrations of Zn occur due to corrosion of household supply system. However, under certain circumstances, tap water can provide up to 10% of the daily intake [29]. Zn is nutritional metal and deficiency in humans has been reported [30-31]. A proposed daily dietary requirement of Zn of 0.3 mg/kg of body weight and a provisional maximum tolerable daily intake (PMTDI) of 1.0 mg/kg of body weight [32] was reported with a daily requirement for adult humans (15–22 mg/day). Its toxicity has been associated with exposure to zinc fumes leading to symptoms of pulmonary distress, fever, chills, and gastroenteritis [27].

2. METHODOLOGIES

The study was designed to follow a two step procedure viz. quality assessment of drinking water samples and explanatory interaction profile of Cd, Pb and Zn on the relative abundance of Fe using multiple linear regression analysis on quality data obtained.

2.1 Drinking Water Sample Collection

Drinking water samples were collected from three locations for this study. They include Gashua (12º52′5″N 11º2′47″E), Monguno (12º41′0″N 13º36′0″E) and Maiduguri (11º50′N 13º09′E). These sampling locations are in the northeastern region of Nigeria. A total of 128 sampling sites consisting 96 ground water (Gashua 32, Monguno 28; Maiduguri 36) and tap water and 32 tap water (Maiduguri only) samples were collected randomly as grab samples (1L) in duplicates from 4 clusters of estimated 5 districts. Sampling technique was as described in Radojevic and Bashkin [33]. Samples were collected in sterile, pre-washed (with detergent, doubly de-ionized distilled water, dilute HNO_3 and doubly de-ionized distilled water, respectively) 500-mL polyethylene bottles. Sampling took place at approximately monthly intervals from April-December 2007. Water samples were obtained directly from the water pumps or taps after allowing the water to run for at least twenty minutes and preserved in acidified to 1% HNO_3 until analysis.

2.2 Sample Analysis

Heavy metals (Fe, Cd, Pb and Zn) concentrations in water samples were determined by Flame Atomic Absorption Spectrophotometry (FAAS) at the NAFDAC Area Laboratory, Maiduguri, using a Shimadzu AA-6800 instrument equipped with ASC-6100 auto sampler. The flame type used for all elements was air-acetylene atomization gas mixture. Water samples were analysed directly according to analytic instrumental procedures [34-35]. Measurements were made using the hollow cathode lamps for the respective heavy metal. Working calibration solutions were prepared by serial dilution from reference standard solutions (1000mg/L) for each metal using 1 % (v/v) HNO_3. A three-point (0.5, 1.0 and 2.0mg/mL) external calibration curve was obtained for the assay of each metal; using the average of three separate readings for each solution. For the determination, each series of analysis included a blank and the calibration curve. Calibration curves were validated by comparisons to recommended quality control parameters for metal analysis [35].

2.3 Data Analysis

Data analysis of results obtained was performed using Analyse-it version 2.26 statistical software for Microsoft Excel. Summary results are presented as mean $\pm$ standard deviation. Decision rule on inferential statistical significance was considered at 95% confidence interval.

The multiple linear regression (MLR) model used in this study expressed the relationship between the three explanatory variables (Cd, Pb; Zn) and the response variable (Fe), by fitting a linear equation to observed data samples. The MLR model equation is:

$$Fe = \beta_0 + \beta_1 Cd + \beta_2 Pb + \beta_3 Zn + \varepsilon \quad (1)$$

where β_0 is a constant; the intercept value, $\beta_{1\text{-}3}$ are the unknown partial regression coefficients of each of the explanatory variables to be estimated, and ε comprises the uncontrolled factors and experimental errors in the model. The least squares fitting was adopted by minimizing the sum of the squares of the vertical deviations from each data point to the line that fits best for the observed data [36-37]. Instances of non-detects and zeros of the explanatory variable were included into the indices and assessed as such due to the possibility of bias into the equation. The variables that showed a non-detectable value may have been due to concentrations in the water samples that were below the detection-limit resulting from either method or instrumental factors [9].

3. RESULTS

3.1 Water Quality Assessment

Mean concentration profile of the metals in drinking water samples analysed from the three locations is shown on Fig. 1. Fe was highest in all locations with remarkable exception of Gashua, where conversely Pb recorded the highest concentration. Fe concentration ranged between 1.317 ± 0.0192 mg/L in Monguno and 0.14 ± 0.003 mg/L in Maiduguri TW. Zn ranged between 0.015 ± 0.008 mg/L in Monguno and 0.007 ± 0.004 mg/L in Maiduguri GW. The mean Cd concentration was maximum in Gashua (0.059 ± 0.001 mg/L) and minimum in Maiduguri (0.004 ± 0.002 mg/L, while the mean Pb concentration ranged between 0.074 ± 0.021 mg/L in Gashua and 0.001 ± 0.001 mg/L in Maiduguri TW. Pb was not detected in all water samples analysed from Monguno.

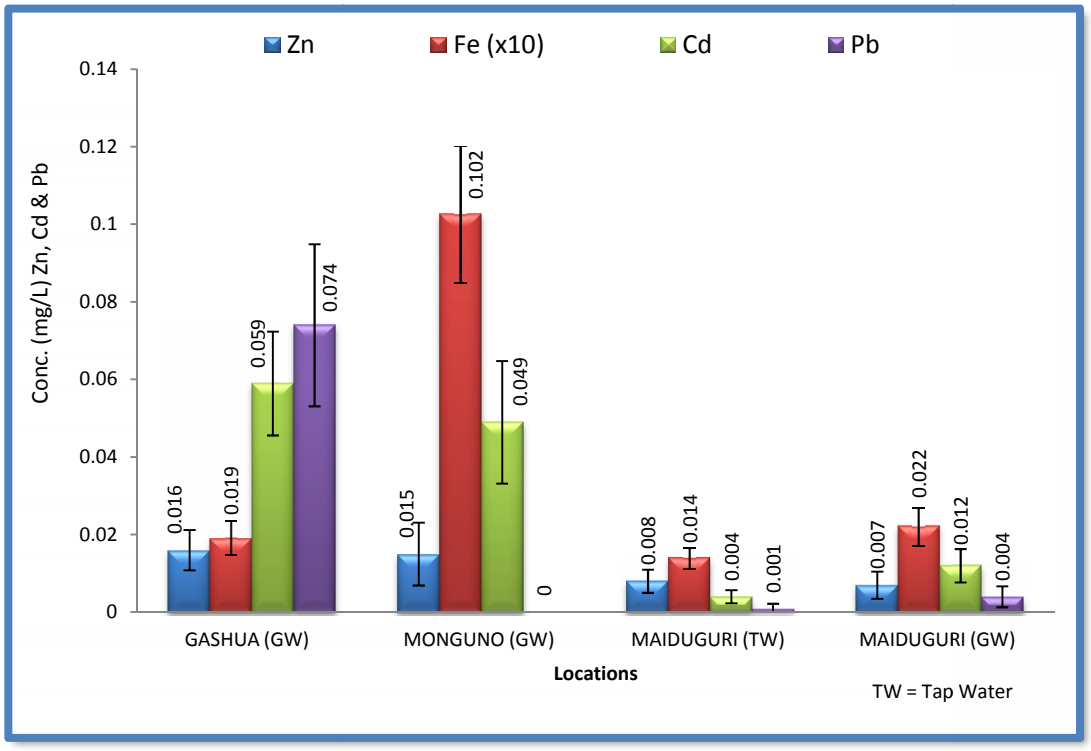

Fig. 1. Mean concentrations profile of metals in drinking water samples

Analysis of variations (1-way ANOVA with Tukey pot hoc test) between each respective metal on the basis of sampling locations statistical significance (P = .05) indicated that Fe concentration was markedly significant in Monguno than other locations, but was not significant between Gashua and all samples (GW and TW) from Maiduguri. This trend was also similarly with Pb concentration in Gashua being significant than all other locations, but was not so between Maiduguri TW and GW. Cd was only significant between Gashua and both Maiduguri TW and GW respectively, and between Monguno and both Maiduguri TW and GW respectively. However no significant Zn concentration variations exist between locations.

3.2 MLR Analysis

Table 1 shows the result of MLR analysis with the regression equation for metals in ground water samples from Gashua, indicating the relationship between the three explanatory variables (Cd, Pb; Zn) and the response variable (Fe). The result indicated R^2 (coefficient of determination) of 0.65, suggesting that 65% of the variations in Fe concentration in water samples from Gashua can be explained by the three metals (Cd, Pb and Zn). The variation is significant. In the regression equation (2), the intercept value (0.068) suggests that the concentration of Fe will be as low as 0.068 mg/L if the concentrations of the explanatory variables are kept constant. Further, the equation revealed that while Cd and Zn are capable of increasing Fe concentration by 1.120 mg/L and 4.172 mg/L respectively, Pb show likelihood of decreasing Fe concentration by 0.057 mg/L. That is, assuming Cd and Zn are held constant, on the average, an increase in Pb concentration variation is likely to influence a reduction in the Fe concentration by about 0.057 mg/L in the ground water system of Gashua.

Table 1. Gashua: MLR analysis result with regression equation for metals in ground water samples

Source of variation	Sum squares	DF	Mean square	F statistic	p-Value	R^2	Adjusted R^2
Model	1.430	3	0.477	17.19	<.0001	0.65	0.61
Residual	0.777	28	0.028				
Total	2.207	31					

Regression Equation: Fe = 0.068 + 1.120**Cd** - 0.057**Pb** + 4.172**Zn** (2)

Table 2 present MLR analysis results of Monguno ground water system. A significant 51% of the variation in Fe concentration in water samples from Monguno can be explained by Cd, Pb and Zn. In the regression equation (3), the intercept value (0.795) suggests that the concentration of Fe will be as low as 0.795 mg/L, although higher than the 0.068 mg/ml of Gashua, if the concentrations of the explanatory variables are kept constant. Similarly, the equation revealed that Cd and Zn are capable of increasing on the average Fe concentration by 3.423 mg/L and 12.210 mg/L respectively, if either metal is kept constant by an increase of the other. Pb was excluded as it was not detected in all samples from Monguno ground water system.

Table 2. Monguno: MLR analysis result with regression equation for metals in ground water samples

Source of variation	Sum squares	DF	Mean square	F statistic	p-Value	R^2	Adjusted R^2
Model	14.059	2	7.029	12.78	.0001	0.51	0.47
Residual	13.746	25	0.550				
Total	27.805	27					

Regression Equation: **Fe** = 0.795 + 3.423**Cd** + 12.210**Zn** (3)

The result of MLR analysis in ground water samples from Maiduguri is shown on Table 3. It indicates an insignificant, low (11%) variation in Fe concentration in the water samples from Maiduguri was explained by Cd, Pb and Zn. The regression equation (4), revealed an intercept value of (0.219) suggesting a possibility of low Fe concentration at 0.219 mg/L, if the concentrations of the explanatory variables are kept constant. This value falls between that obtained in Gashua and Monguno. The regression equation (4) revealed that only Cd has potential incremental influence on the average Fe concentration, while Pb and Zn posed as agents of reduction in Maiduguri ground water system.

Table 3. Maiduguri (GW): MLR analysis result with regression equation for metals in ground water samples

Source of variation	Sum squares	DF	Mean square	F statistic	p-Value	R^2	Adjusted R^2
Model	0.328	3	0.109	1.34	.2778	0.11	0.03
Residual	2.601	32	0.081				
Total	2.929	35					

Regression Equation: **Fe** = 0.219 + 3.577**Cd** - 3.249**Pb** - 2.913**Zn** (4)

Table 4 shows the MLR analysis results of Maiduguri tap water system. Similar to the ground water results, it indicates an insignificant, low (17%) variation in Fe concentration in the water samples explainable by Cd, Pb and Zn. Again the regression equation (5), revealed a similar trend of low Fe concentration at 0.200 mg/L, if the concentrations of the explanatory variables are kept constant, as in the Maiduguri ground water. This value falls between that obtained in Gashua and Monguno. The regression equation (5) show that all explanatory variables have potential decreasing influence on the average Fe concentration in tap water system from Maiduguri.

A comparative trend of Fe responses to the explanatory interaction profile of metals in drinking water samples at the different locations is graphically summarized on Fig. 2. In addition to the foregoing, the following general conclusion can be drawn from the MLR analysis:

- Pb is a key agent of reducing Fe concentration variations in water systems and the impact tend to be more rigorous when the concentrations of the Cd and Zn are comparatively higher than it.
- Cd and Zn tend to influence increases in Fe concentration variations in water systems, but Zn immensely confer this influence in the absence of Pb

Table 4. Maiduguri (TW): MLR analysis result with regression equation for metals in tap water samples

Source of variation	Sum squares	DF	Mean square	F statistic	p-Value	R^2	Adjusted R^2
Model	0.128	3	0.043	1.94	0.1462	0.17	0.08
Residual	0.618	28	0.022				
Total	0.747	31					

Regression Equation: Fe = 0.200 - 2.527**Cd** - 3.083**Pb** - 1.561**Zn** (5)

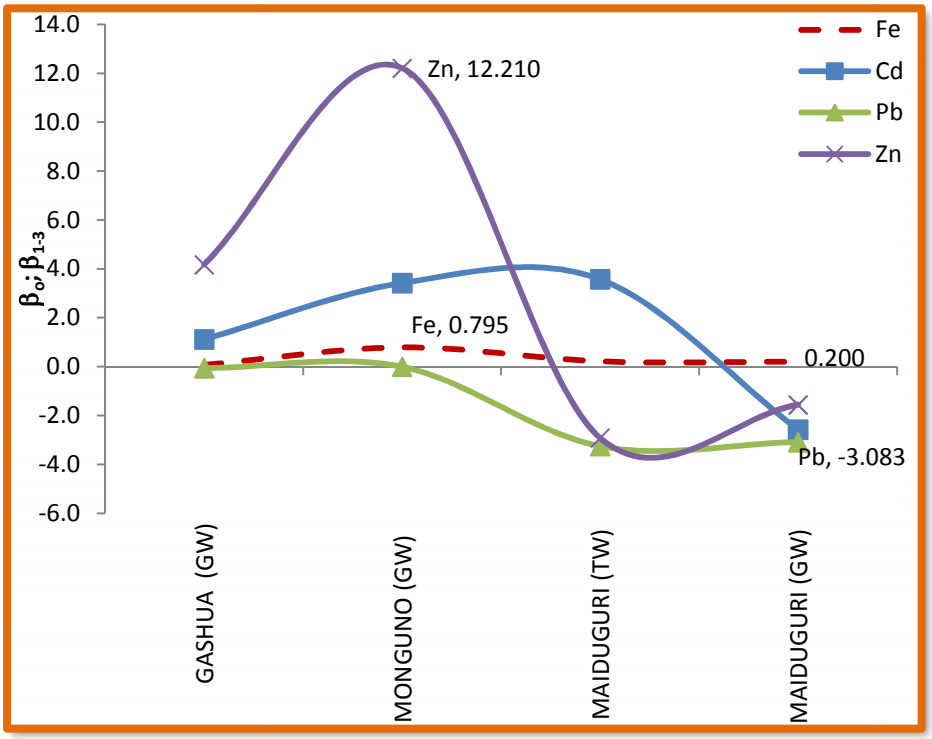

Fig. 2. Graphical trend of Fe response to the interaction profile of metals in drinking water samples at the different locations

4. DISCUSSION

In Nigeria, ground water from hand dug shallow wells and bore holes (deep well) are the major source of drinking water to more than 70% of the population [38]. Therefore continuous monitoring ground water sources of drinking water are imperative.

The widespread variations in concentration of metals determined from the region may be attributed to the depth of the boreholes sampled. The depth in Gashua, Monguno and Maiduguri were 70.0, 371.5 and 454.5 meters respectively. These mean depths fall within the upper, middle and lower source aquifers which exist in the Chad Basin that are assumed to be typically used for potable ground water supply in the region [39]. Rainfall dilution effect is another factor especially in Maiduguri where the upper and middle aquifers have been

exploited intensively [39-40]. Thus the results are clearly defined by the source aquifers and consistent with the current extent of metal contaminations of the aquifers in the Chad Basin region [41-42]. The shallow upper aquifers are prone to contamination but the underlying aquifers are protected and relatively free of toxic elements [43].

Pb was not detected in Monguno samples. This could be the result of shared geological cycling of Cd, Pb and Zn [44]. Cd shows slight enrichment in clayey zones rich in soluble Fe-hydroxides while Pb shows little or no association with Fe-Mn precipitates. Thus, the high levels of Fe, Cd and Zn recorded in the Monguno water samples could be the consequence of the generally high element content of the region's soil rather than an indication of pollution as reported elsewhere [45]. Similarly, the low element contents of Maiduguri tap water could be as a result of low human activities around the dam site and the treatment carried out before distribution to end users.

According to the WHO guidelines [23], Cd and Pb show potential implications for public health especially in Gashua. However the result of this study shows concordance with earlier findings in the upper aquifer within the Chad Basin [41-42] and in other parts of Nigeria. Such as the from hand-dug wells and shallow boreholes in Zaria metropolis, North Central Nigeria [46], Akure, South Western Nigeria [47] and overseas [48-49]. Generally, in areas where good quality management systems are in place the levels of toxic elements found in drinking water, whether in its natural state or after treatment, are always below the permissible maximum levels stipulated by the World Health Organization and the Water Pollution Control Regulations of the respective countries [49].

Zn is found naturally in water [26] most frequently in areas where it is mined and it enters the environment from industrial waste, metal plating or sludge. The later reasons are the case with the current study. However the range of Zn in this study compares with findings from other parts of Nigeria [50]

Fe was beyond guideline levels only in Monguno. On the contrary a number of studies have noted the occurrence of high Fe concentrations in confined ground waters of the sedimentary basins. Higher concentrations of total Fe were reported in ground waters from sedimentary aquifers in southeastern Nigeria [51-52], South-West Nigeria [47] and the Niger Delta [53]. In the Chad Basin, middle aquifer Fe levels of up to 20 mg/L have been found [41-42].

The MLR analysis on explanatory interaction profile of Cd, Pb and Zn on the relative abundance of Fe as response variable in this study buttressed the literature findings concerning the depth of aquifers in the Chad Basin region [41-42], geological cycling [44] and importantly the chemical nature of the metals [3,10,22;26] releases due to ore formations in ground water systems [3]. For instance the high (65%) R^2 in Gashua confirms of existence of sulfide ore in this region.

4. CONCLUSION

This study obtained and presented the result of quality status of drinking water in three locations of northeastern Nigeria showing the need for, specifically Cd and Pb, Contaminant Control Programmes in ground water from Gashua.

In the attempt to answer the question: How does the relative abundance of Fe vary as the interact profile of Cd, Pb and Zn varies in drinking water quality status? The findings of the

MLR analysis show that Fe varied diversely due to difference in the concentrations of these metals in the aquifer locations of all three explanatory variables. Fe was found to be potentially lowered by the shared presence of all three explanatory variables, but was greatly influenced by the presence of Pb.

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COMPETING INTERESTS

Authors have declared that no competing interests exist.

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