Assessment of Heavy Metal Distribution in Surface Water and Groundwater in The Region of a Salt Mine

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ABSTRACT: *Water quality is a big concern for the humankind as it is the most important natural resource. The quality of water is affected by anthropogenic activities carried which could render it unsuitable for human consumption. The present study assessed heavy metal distribution and toxicity in surface and groundwater resources of an area characterized by salt mining from brine ponds. Samples were analyzed for pH, electrical conductivity (EC) elemental compositions of: Ba, Mn, Fe, Cu, Sr, and Zn to assess their spatial distribution, sources, variability, toxicity and possible health risks. Median concentrations of Ba, Mn, Fe, Cu, Sr, and Zn in surface water was: 0.28, 3.63, 2.23, 0.03, 0.14 and 0.05 respectively while for groundwater, median concentration was: 0.65, 0.20, 1.16, 0.03, 0.83 and 0.11 respectively. In terms of suitability, concentrations of Mn, Fe and Sr in surface water, and that of Sr, in groundwater raises some quality concerns since they are all present in concentrations above the permissible limits for drinking water. Results of Corelation and Principal Component Analyses showed that source and mobility of these metals is linked to both geogenic (host rock weathering) and anthropogenic activities mainly associated with salt mining and processing in the region of a saline pond. Spatial distribution of concentration of these metals also shows higher concentrations in the immediate region of the salt mine especially in groundwater.*

KEYWORDS: Heavy metals, Salt mining, Water quality, correlation analysis, principal component analysis

INTRODUCTION

Surface water and Groundwater constitute the major sources of water, a resource that is essential for sustenance of all living organisms (Bytyci *et al.,* 2018). The distribution of freshwater resources is unequal across the globe, and freshwater availability is becoming increasingly scarce as a result of population expansion and varied human activities These sources of water can be affected by poor management practices as well as anthropogenic activities that may release pollutants into the

environment. Physico – chemical characteristics of all waters reflect processes present within the water cycle that are natural processes or as a result of anthropogenic activities. In the absence of fresh precipitation, surface water and groundwater are used to satisfy the demands of diverse industries.

Heavy metals are metals and metalloids that are toxic to organisms at high concentrations, and their accumulation threatens the quality of both surface water and groundwater and can accumulate in the food chain (Ali et al., 2019). Exposure to these metals can lead to physical, muscular and neurological disorders (Ali et al., 2019), while long-term exposure can also cause deadly diseases like Alzheimer's, Parkinson's, multiple sclerosis, and cancer (Mishra et al., 2010). The assessment of the quality as well as heavy metal load of surface and groundwater for any purpose, notably for human consumption and the spatial and temporal variation in the quality water sources in response to local geologic set-up and anthropogenic influences is necessary.

Heavy metals can be mobilized by salts such as sodium chloride (NaCl), magnesium chloride ($MgCl_2$), and calcium chloride ($CaCl_2$) into water bodies (Schuler and Relyea, 2018) since in most cases, they are not readily available i.e., bound to rocks and soils. Salts can transform metals to free ions, soluble species, and weakly adsorbed species (Violante et al., 2010) which ultimately increases their concentrations in such resources. Salt is generally produced by one of three ways: deep-shaft mining, solution mining or solar/artificial evaporation. In each case, salt is washed, drained, cleaned and refined; surface and ground waters may be affected by discharges of contaminated water often loaded with salt from these processes (Goldan *et al.,* 2014).

Inland brines are known to occur within continents as opposed to coastal regions. The study area is centered on Keana Town, Central Nigeria. This town is popular for mining of brines to produce salts; the brines are known to occur in ponds that appear to originate from underground brines issuing from the western flanks of Keana anticline and the interbedded shale of fractured sandstone of Awe Formation in the Central Benue Trough of Nigeria (Offodile, 1984). Sallau *et al.,* (2016) in a study that involved analysis of trace element concentration in soils of the study area discovered an enrichment in the elements: Mo, Co, Cr, Cu, Ba, Ni, As, and Zn. The sediments are therefore contaminated by these metals and may in turn affect surface and groundwater quality as leaching of these metals is made possible by runoff containing salts from salt processing activities or simply due to precipitation. The impact on these water sources can have negative impacts on health and wellbeing (Christophoridis et al., 2009). The presence of brine constitutes a serious hydrogeological problem on groundwater and as noted by Adeoti et al. (2010), saltwater intrusion into aquifers has become a major concern in most of the areas around brine fields as it constitutes the commonest of all the pollutants in freshwater. According to Winter *et al.,* (1998), groundwater - surface water interaction has a major role in affecting chemical and biological processes in surface water bodies and water quality of the hydrological system is therefore influenced if variables such as pH, temperature and dissolved oxygen are altered.

The study area is characterized by a tropical sub-humid climate with two distinct seasons: dry and wet. The dry season starts from November to April, and the wet season is from May to October. The mean annual rainfall ranges from 1000mm to 1500mm while the mean annual humidity is 70% and relative humidity 60% to 80% (NIMET, 2020). The mean annual temperature is 28.5⁰C with mean annual sunshine of 7 hours per day. A high temperature of 33⁰C to 36⁰C is experienced in the areas during the dry season (NIMET, 2020). The study area has a low to moderate relief with few scattered laterite capped hills of elevation ranging from 100m–160m above mean sea level. The drainage system which shows dendritic pattern is drained by minor tributaries of the River Benue such as River Ome, River Akinakwa, River Bid, River Alashami, River Okpalaga and River Okpikpi, although most of the rivers are seasonal rivers. The study area forms part of the Keana brine field situated in the Middle Benue Trough (Benkehlil, 1986). Main rock types are argillaceous and of continental and marine origin dominated by sandstone with traces of shale. The sandstone is indurated, medium to fine grain size and felsic in nature, while the shale is fissile, which is believed to be an extension of the Awe Formation.

The present study aimed at assessing the heavy metal distribution in the region of a salt mine in both surface and groundwater using elemental concentrations and statistical analysis.
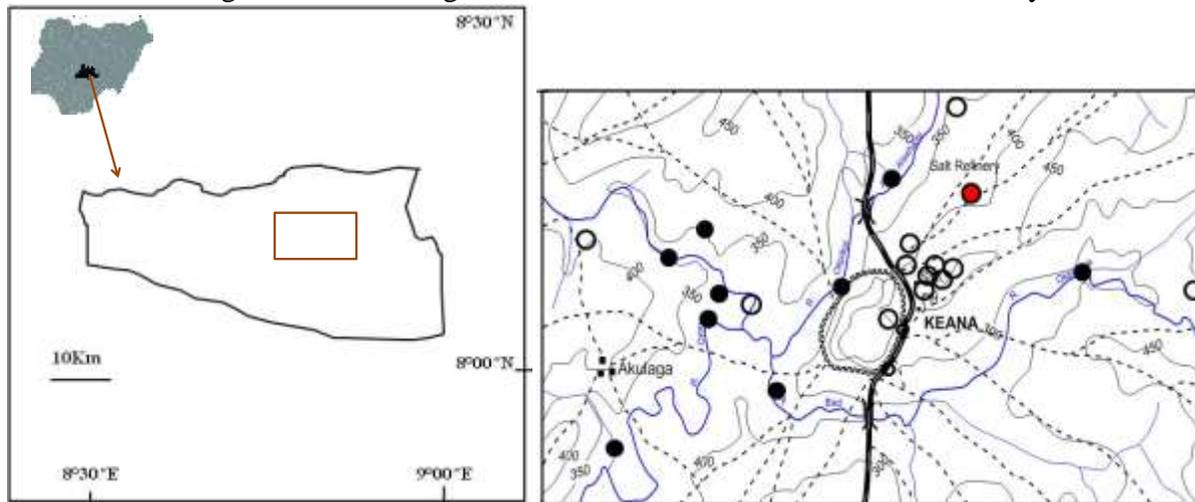
Figure 1. Location of the study area, also shown is relief and drainage in the study area as well as sample collection points.

METHODOLOGY

Water Sampling
Water samples were collected at intervals along the major drainage in the study area and its tributaries. Groundwater samples were collected form water wells/boreholes as close as possible to surface water collection points (figure 1). High density polyethylene bottles were properly rinsed (three times) with acid to collect the samples. A total of twenty-five water points were

assessed: sixteen groundwater and nine surface water points. Water level measurements were made in hand dug wells and where possible in boreholes. pH, Electrical Conductivity (EC), and Temperature (T) were measured *in-situ*, while samples were taken for further geochemical analysis in the laboratory.

Water samples collected from each point were transferred into sterile 120ml containers and subsequently acidified with 2 to 3 drops of HNO_3. The samples were shipped to Activation Laboratories Limited, Ontario, Canada to analyze for elemental concentration. The elements analyzed are Ba, Al, K, Mg, Mn, Ca, Fe, Cu, Na, Sr, and Zn, using Inductively Coupled Plasma-Optical Emission Spectrometry (ICP-OES).

Data Analysis
Available data was values of physical parameters measured *in-situ* i.e., pH, T and EC and concentrations of elements in the water samples. Preliminary treatment of data involved inspection and identifying detected elements with respect to detection limit of the instrument used. This was followed by a test for normal distribution after which measures of central tendency were done. For the major cations, a bar plot of the median concentration for sample point in each case of groundwater and surface water was done to establish their relative abundance. Gibbs diagram was used to identify the origin(s) of these cations in the water samples. These were all done using Microsoft Excel Software (365).

For the heavy metals, box plots were used to assess and identify their relative abundance and to compare the concentration in water to published standards mainly the Nigerian Standard for Drinking Water Quality NSDWQ (SON, 2007). Surface plots of the trace element concentration was produced using MATLAB; this was done to establish the variability in their concentration and attempt to link this with activities within the study area, particularly salt mining around the saline pond. Trace element concentrations in water were statistically analyzed using the statistical toolbox in MATLAB (Rb2007). The median concentrations and standard deviations of the elements in water were calculated. Correlation analysis among heavy metals was performed using Pearson's coefficient (with $p < 0.05$ considered as statistically significant). Principal Component Analysis was used to identify potential sources of these heavy metals in the water samples.

RESULTS AND DISCUSSION

Character of hydro-chemical indicators
Groundwater had pH values ranging from 7.5 to 8.0, while that of surface water ranged from 7.1 to 8.0 all indicating alkaline conditions, EC values varied from; 15µS/cm to 144µS/cm in groundwater, while the range in surface water is 10µS/cm to 24µS/cm. EC and TDS are relatively higher in groundwater because of leaching from soil or from the interaction of different sources of groundwater, while temperature is similar for both water sources. Water from the salt pond as

expected has higher EC, TDS and Temperature values than surface and groundwater samples. Water sample from the salt pond was warmer (38.2°C), had a higher TDS value: 11375µS/cm and a more alkaline pH: 8.7. EC values are indicative of water quality in the areas unaffected by salinity; thus, the low EC value recorded in both surface and groundwater in the study area indicates better water quality than that of other surface waters.

Coefficients of skew for major cations and heavy metals in the water samples showed that concentrations of the elements were not normally distributed, i.e., >0 (table 1a and 1b); the median values of concentrations of the heavy metals were used to assess the two sources of water instead of the mean values (Krishna *et al.,* 2009). The order of relative abundance of major cation in the surface water is Na>Ca>K>Mg>Al. The order of relative abundance of major cations in the groundwater is Ca>Na>K>Mg>Al.

Table 1. Coefficients of Skew for elemental concentration in Groundwater and Surface Water

Element	Al	K	Mg	Ca	Na	Ba	Mn	Si	Fe	Cu	Sr	Zn	Al
Surface Water	1.62	0.29	0.23	0.25	0.10	0.22	3.42	0.32	1.79	1.30	0.40	3.23	1.62
Ground Water	0.10	-0.12	0.65	1.07	1.57	1.90	1.33	-0.40	-0.06	-0.26	1.15	1.44	0.10

The origins of major cations are determined using Gibbs plot. Gibbs (1970) suggested; TDS versus Na/ (Na + Ca) for cations to illustrate the natural mechanism controlling water chemistry, including the rainfall dominance (precipitation), rock weathering dominance (water rock-interaction), and evaporation dominance.

Gibbs diagram (figure 2) shows that 87% of the groundwater samples fall within water-rock interaction dominance field, while 13% fall within the precipitation dominance field. Surface water samples plot predominantly within the precipitation dominance field, while 33% fall within the water-rock interaction dominance field. From the percentages it can be deduced that the dissolved solids are derived from interaction with the host rocks and more so in groundwater that has enough residence time to allow for such interaction. Precipitation being the main source of recharge to both surface and groundwater sources will have some influence on the TDS content; TDS in surface water samples is derived mainly from precipitation, while few where derived from combination of both host rock and rainfall.
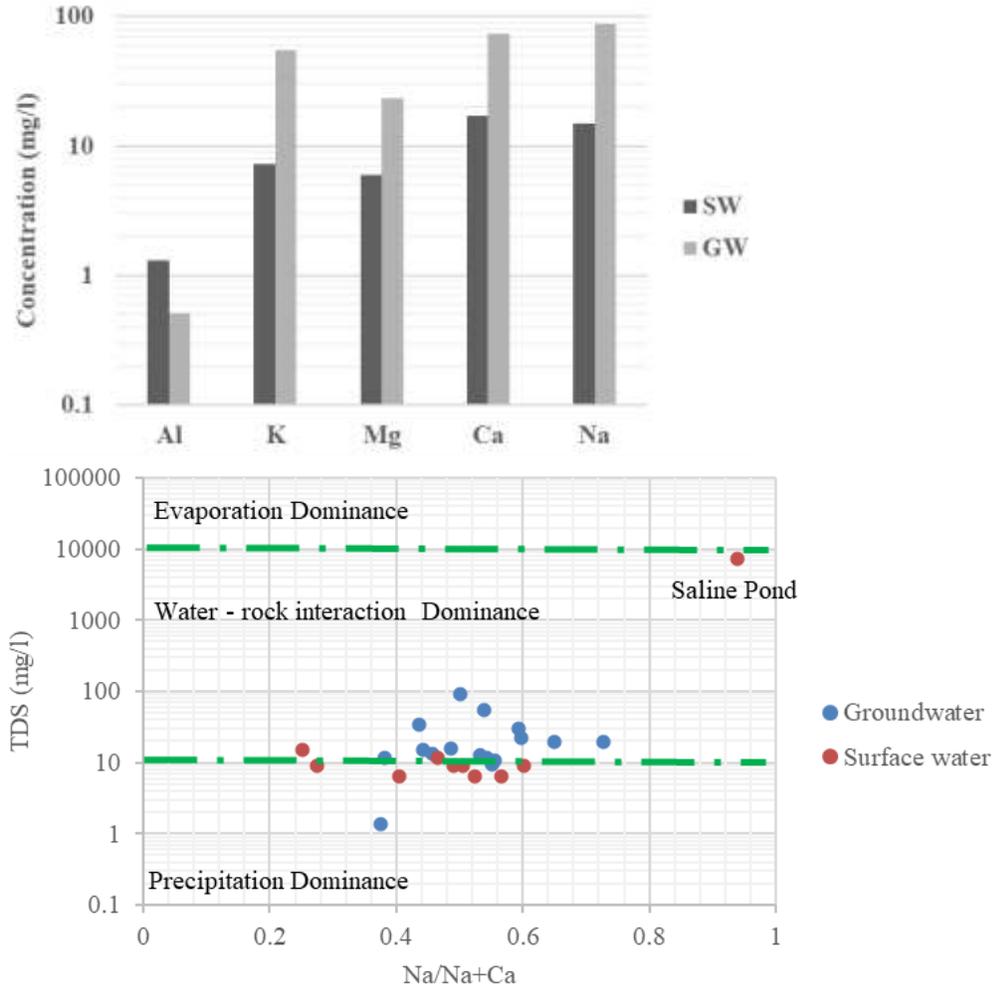
Figure 2. Relative abundance of major cations in surface and groundwater (top);Gibbs diagram showing the major contribution to cation load in surface and groundwater (bottom).

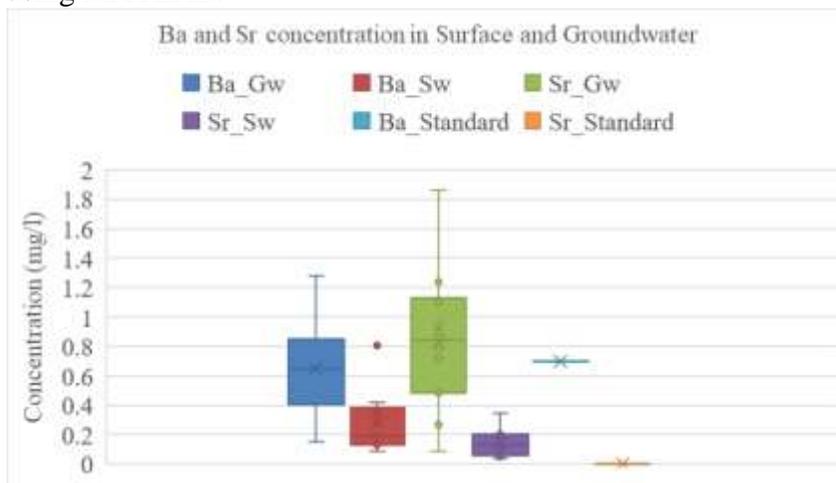***Heavy metals in surface and groundwater sources***
The distributions of Ba, Fe, Mn, Se, V and Zn and their parameters in surface water and groundwater are presented as box plots. The bottom and top of the box represents the 1st and 3rd Quartiles (Figure 3). Relative abundance of the heavy metals analyzed for in groundwater is in the order: Sr>Ba>Fe>Mn>Zn>Cu; while that of surface] water is in the order: Fe>Mn>Ba>Sr>Zn>Cu. The two sources show different order of abundance of these heavy metals. Maximum concentration of Ba in groundwater is 1.28mg/l and is higher than that recorded in surface water i.e., 0.8mg/l. maximum concentration of Sr in groundwater (1.86mg/l) also exceeds the concentration in surface water (0.35mg/l) and the permissible limit for drinking water purposes. Same is the case also for Zn and Cu, concentration in groundwater (0.25 and 0.07mg/l respectively) is higher than that of surface water (0.088 and 0.04mg/l respectively) although the concentration

in both cases is within permissible limits for drinking water purposes. Maximum concentration of Fe and Mn is however higher in surface water (4.27 and 11.9mg/l respectively) than in groundwater (1.67 and 5.39mg/l respectively). Concentration of Mn in surface water and Fe in both surface and groundwater exceed permissible limit for drinking water purposes. In terms of suitability, concentrations of Mn, Fe and Sr in surface water, and that of Sr, in groundwater raises some quality concerns since they are all present in concentrations above the permissible limits for drinking water according to the Nigerian Standard for Drinking Water Quality (SON, 2007).

Spatial distributions of the heavy metals are shown in figures 4a and 4b. Spatial plots for concentration of Ba, Cu and Sr in groundwater show similar patterns with highest concentration around the salt pond/salt mining region (figure 4a). concentration of Mn and Fe also decrease away from the region. High concentrations of these elements in groundwater are also seen in the region of the main township settlement likely associated with poor sewage disposal methods. Spatial plots for concentration of Fe, Cu, Ba and Mn in groundwater also display a similar pattern, highest concentration is recorded along the drainage channels. The pattern exhibited by Sr in surface water is such that the concentration is high in the region of the salt pond/mine.

Statistical Analysis on trace element concentration
The data were statistically analyzed by using the statistics toolbox in MATLAB Rb2007c(Mathworks). Correlation analysis among the elements was performed using Pearson's coefficient. Multivariate methods in terms of principal component analysis (PCA) were used to infer the potential sources of the heavy metals in both surface and groundwater (Liang et al. 2015). The Eigenvalues were used as the extraction method to find out the principal components (PC) during PCA analysis. Surface maps of the spatial distribution of the heavy metals were prepared using MATLAB.
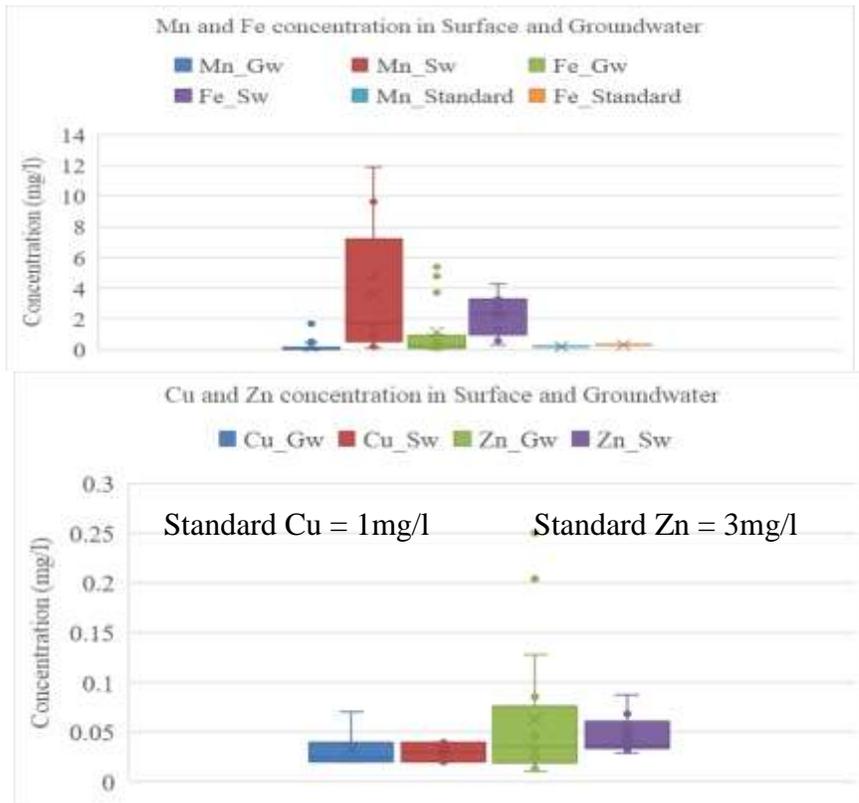
Figure 3: Heavy metals in Surface and Groundwater; also plotted is the Permissible Limit (all NSDWQ except Sr which is USEPA) of these Elements in Drinking Water.

Statistical analyses were performed to reveal the associations and identify relevant factors that control the dispersion of these elements in both surface and groundwater. According to Proshad et al., (2019) interactions between heavy metals can indicate their sources and pathways for dispersion. For the present study, inter relationships between the elements and possible sources were determined using Pearson's correlation coefficient *r*. Pearson's correlation coefficient values between 0.9 and 1 are strongly correlated, while values ranging from 0.9 to 0.5 are moderately correlated (Tirkey et al. 2017). The present study also incorporated this classification into the analysis to have an overall idea about the contribution performance of measured water quality parameters. For groundwater, statistically significant correlations were observed between Ba and Cu, Ba and Sr (r = 0.79 and 0.74 respectively); between Mn and Fe (r=0.75) and Mn and Sr (r=0.64). In surface water, correlation was observed between Ba and Mn and Ba and Sr (r = 0.88 and 0.90 respectively); Mn and Sr (r = 0.64), Fe and Cu (r = 0.75), EC and Fe (r = -0.50) and EC and Cu (r = 0.58). Considering the relationships between the combinations exhibiting significant positive associations indicated that the parameters were interrelated with each other and maybe originated from the same sources in the study area (Abbasi et al. 2013).
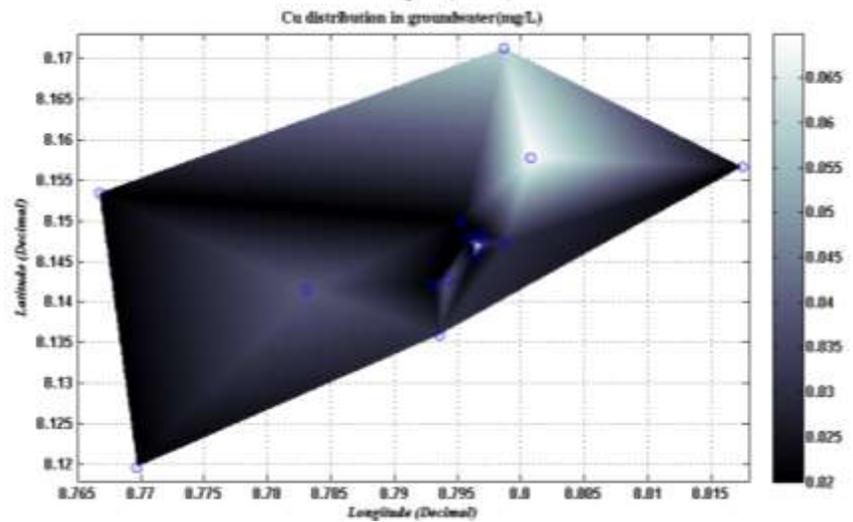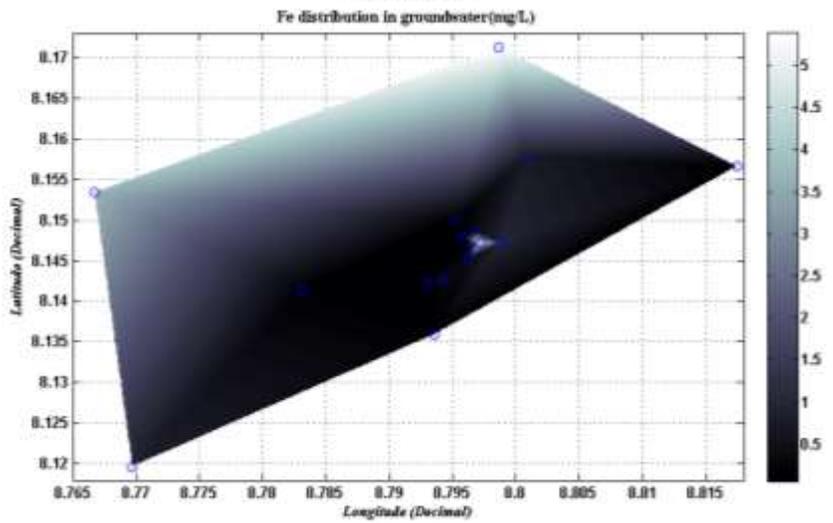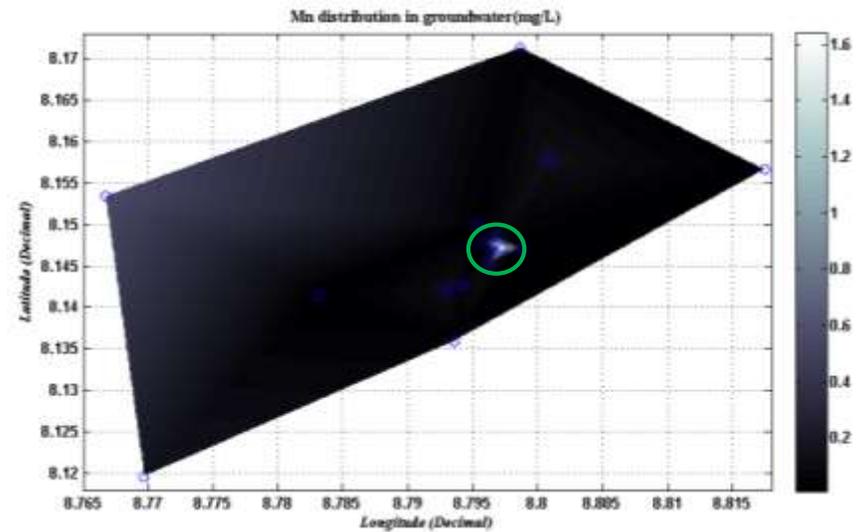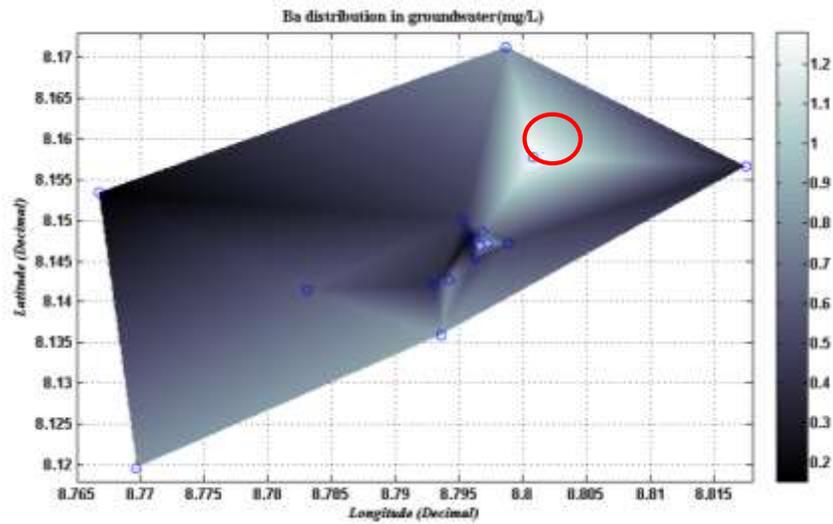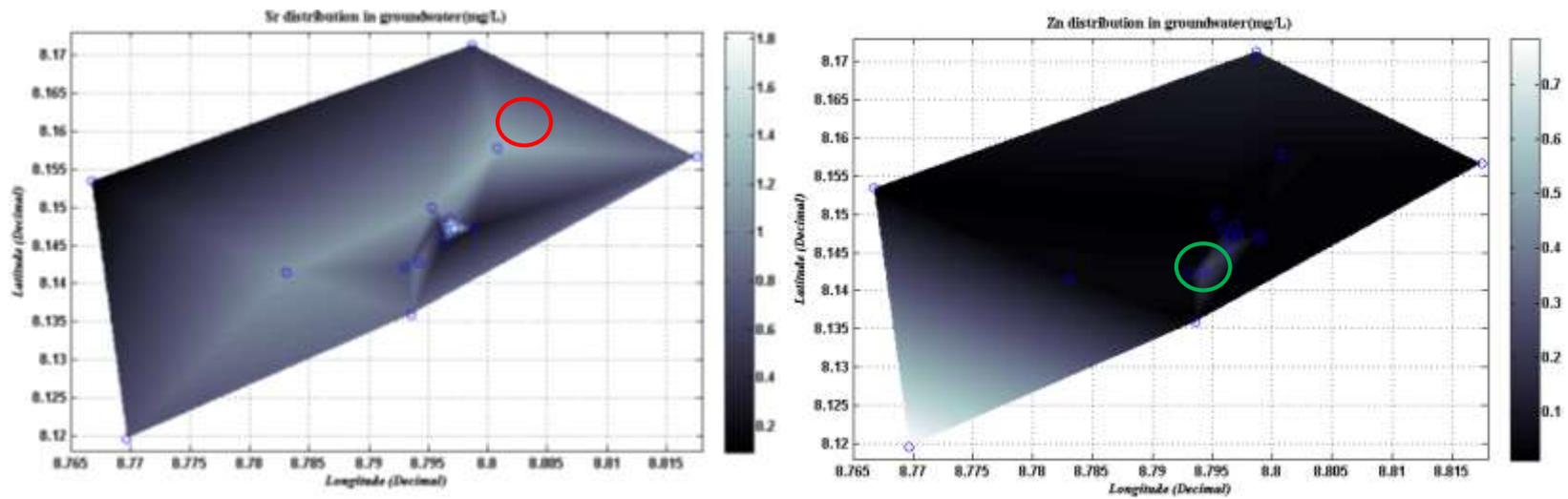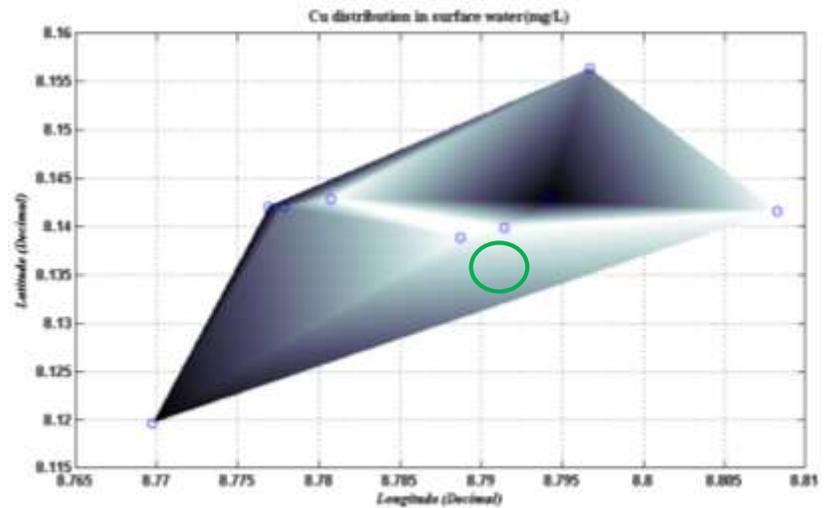
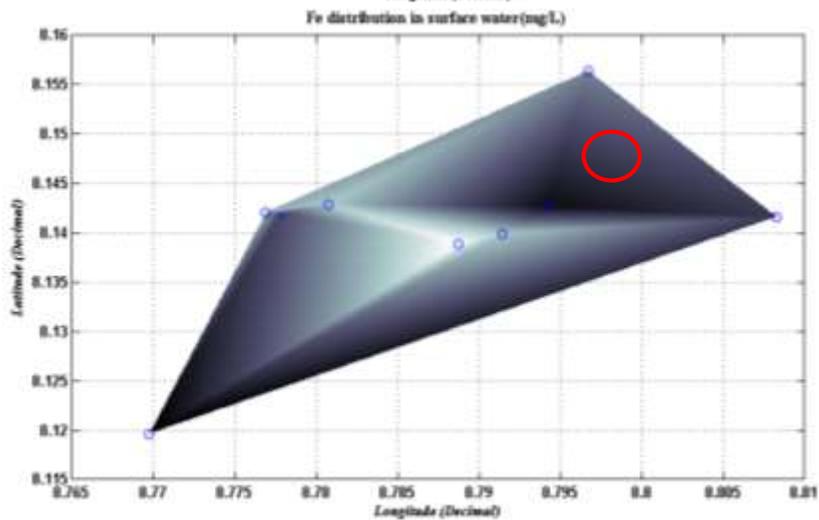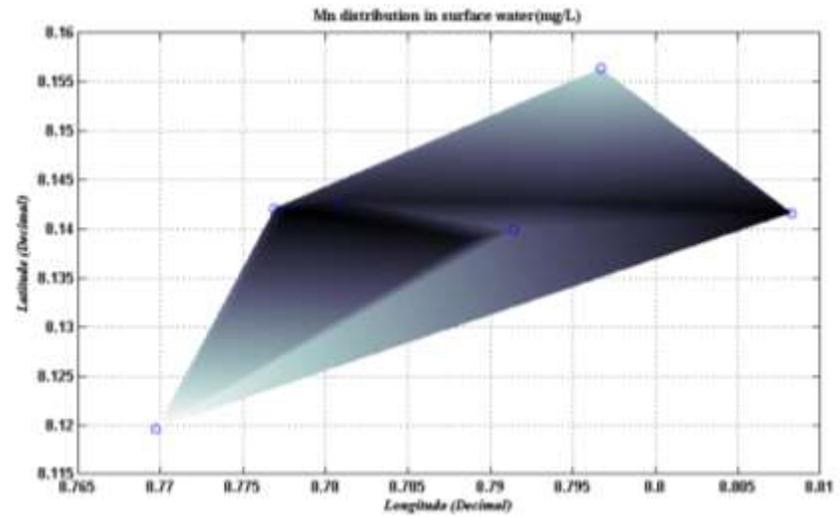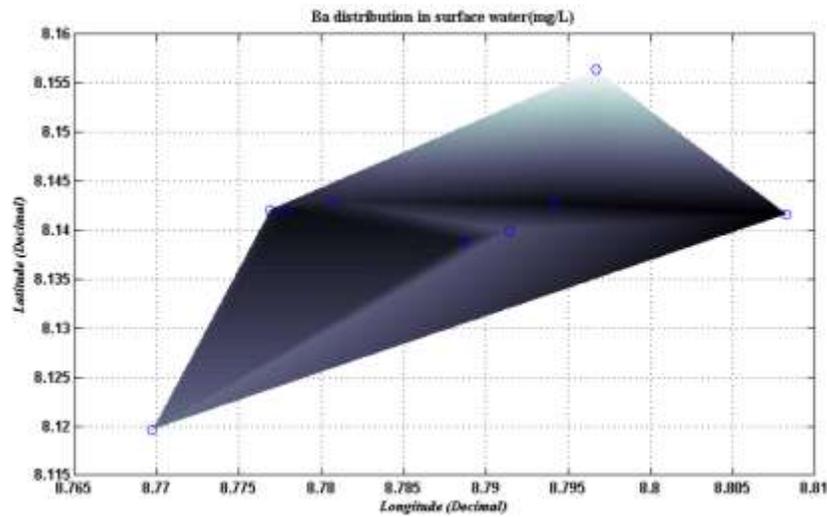Figure 4a. Spatial distribution of heavy metals in groundwater. Red circle indicates region of the salt pond and salt mining activities, while the green circle indicate region og the main town settlement.
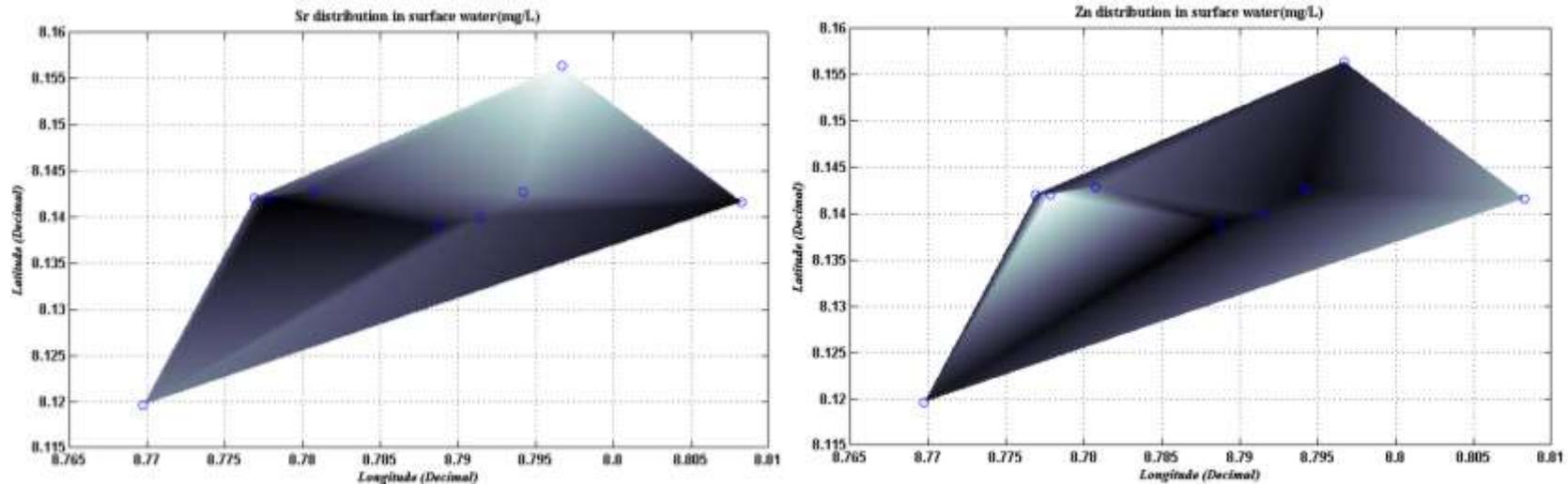
Figure 4a. Spatial distribution of heavy metals in surface water. Red circle indicates region of the salt pond and salt mining activities, while the green circle indicate region og the main town settlement

Table 2. Pearson's correlation coefficient matrix for groundwater (top) and surface water (bottom)

	Ba	Mn	Fe	Cu	Sr	Zn	EC
Ba	1.00	0.88	0.29	0.11	0.90	0.12	0.07
Mn	0.88	1.00	0.01	-0.13	0.83	0.01	0.48
Fe	0.29	0.01	1.00	0.67	0.09	0.11	-0.50
Cu	0.11	-0.13	0.67	1.00	-0.18	0.35	-0.58
Sr	0.90	0.83	0.09	-0.18	1.00	-0.04	0.23
Zn	0.12	0.01	0.11	0.35	-0.04	1.00	-0.08
EC	0.07	0.48	-0.50	-0.58	0.23	-0.08	1.00
Ba	1.00	0.44	0.35	0.79	0.74	0.31	0.42
Mn	0.44	1.00	0.75	0.25	0.64	0.25	-0.06
Fe	0.35	0.75	1.00	0.40	0.27	0.28	-0.11
Cu	0.79	0.25	0.40	1.00	0.49	0.34	0.44
Sr	0.74	0.64	0.27	0.49	1.00	0.11	0.23
Zn	0.31	0.25	0.28	0.34	0.11	1.00	-0.05
EC	0.42	-0.06	-0.11	0.44	0.23	-0.05	1.00

Other relationships were not statistically significant while the positive relationship between Ba, Cu, Mn and Sr in both groundwater and surface water indicating similar sources and mobility in both cases i.e.: geogenic inputs. These elements may have been leached from weathered rocks in the study area. On the other hand, there exists and insignificant correlation between EC and the heavy metals except in the case with Fe and CU in surface water. The lack of any correlation with the elements in groundwater implies that EC is not a main factor affecting the mobility in the water (Barzegar et al., 2016). Correlations between EC and Fe and EC and Cu indicated mutual dependence, common sources and identical behavior of their influence (Jiang et al. 2014).

Principal component analysis (PCA) is an effective tool for source identification of heavy metals in surface and groundwater (Proshad *et al.*, 2020; Zhou *et al.*, 2020) as such the same analysis to aid in identification of possible sources of the heavy metals studied. PCA analysis incorporated the data on concentration of the six heavy metals in both surface water and groundwater. Outcomes of the PCA for surface and groundwater are presented in figure 5. The Principal Component 1 (PC1) usually accounts for the highest variability in any dataset measured (Emenike et al. 2018). In this study, six PCs were computed, and the variances explained by them were 92.08%, 7.86%, 0.05%, 0.01%, 0.00%, and 0.00% for surface water. For groundwater the variances were: 90.12%, 7.43%, 1.77%, 0.5%, 0.11%, and 0.00%. The first two principal components (PC1 and PC2) had Eigenvalues greater than one and are thus considered the most significant.
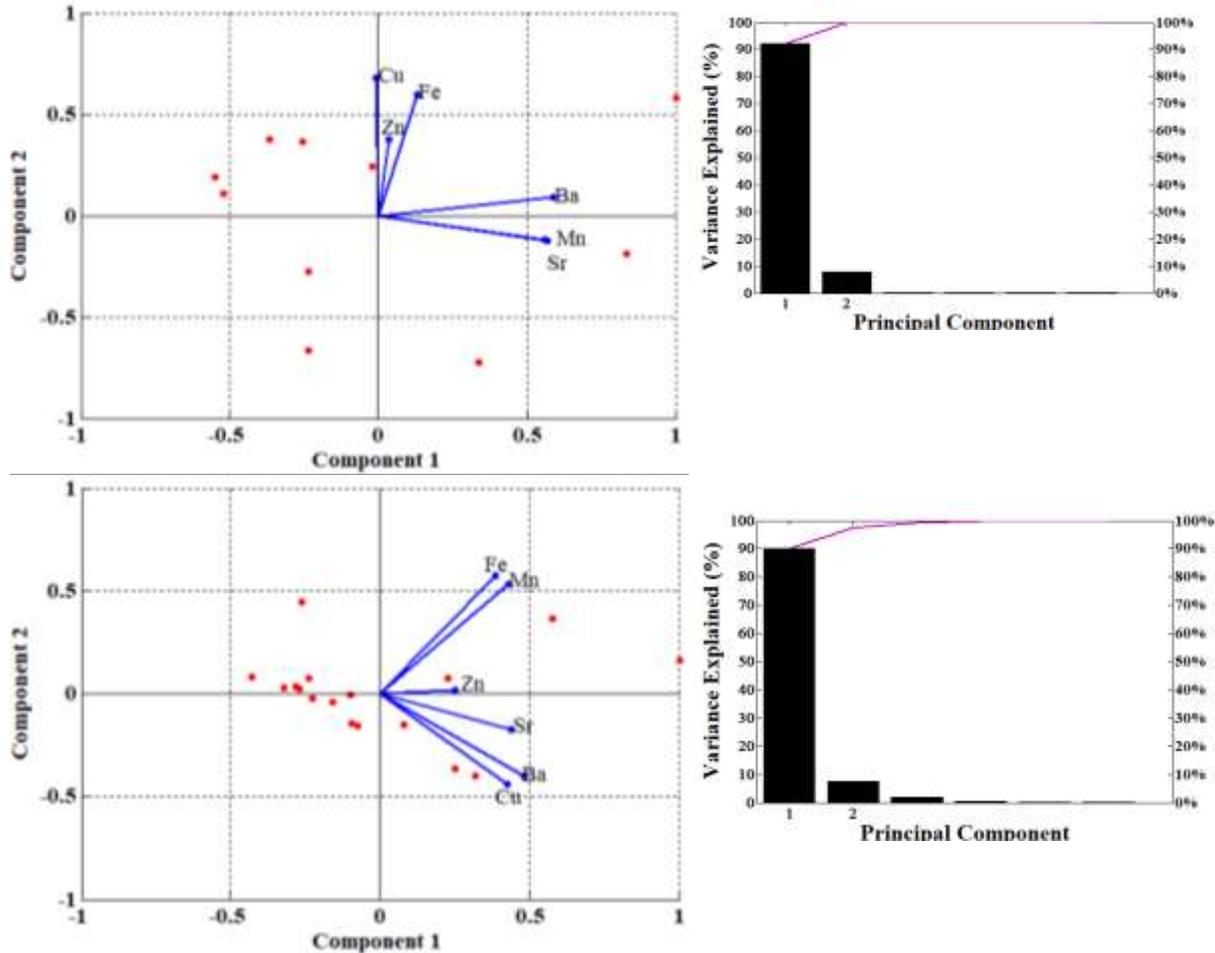
Figure 5. Biplots (and scree plots) of PCA results for trace metal concentration (variability) in surface water (top) and groundwater (bottom).

For surface water, PC1 accounts for 92.08% of the total variance (figure 5) and is dominated by the strong factor loadings for Ba, Mn, and Sr, while moderate factor loading for Fe and Zn and low factor loading for Cu. PC2 accounts for 7.86% of the variance (figure 5) and is dominated by Cu and Zn having strong factor loadings; it also has a moderate factor loading for Fe. The moderate loading for Fe in both the PC1 and PC2 is indicative of its predominance as a pollutant in surface water in the study area.

For groundwater samples, PC1 accounts for 90.12% of the total variance (figure 5) and is dominated by the strong factor loadings for Cu, Ba, Zn, and Sr, while moderate factor loading for Fe and Mn. PC2 accounts for 7.43% of the variance (figure 5) and low to moderate factor loading for Fe and Mn. The moderate loading for Fe and Mn in both the PC1 and PC2 is indicative of their predominance as pollutants in groundwater of the study area.

Based on correlation coefficients and PCA, the six heavy metals investigated can be grouped into two main clusters in surface water i.e., Ba-Mn-Sr and Cu-Fe-Zn. Groundwater samples can be grouped into three clusters i.e., Cu-Ba-Sr; Zn; and Fe-Mn. These clusters have probable common sources and controlling factors for mobilization, could have been mobilized by salt bearing wastewater from the salt mines. The associations can also be reflective of a mixed process where natural weathering releases the elements which are then remobilized acts in addition to anthropogenic factors. This clearly indicates both natural and anthropogenic influence on both surface and groundwater with respect to the studied heavy metals.

CONCLUSIONS

The present study assessed the trace metal content of surface and groundwater in the region of a salt mine. The main conclusions of this study are: (1) Major cations in surface water are sourced from precipitation while that in groundwater are sourced from water-rock interaction. The order of relative abundance of major cation in the surface water is Na>Ca>K>Mg>Al; while for groundwater is Ca>Na>K>Mg>Al. (2) In terms of suitability, concentrations of Mn, Fe and Sr in surface water, and that of Sr, in groundwater raises some quality concerns since they are all present in concentrations above the permissible limits for drinking water according to the Nigerian Standard for Drinking Water Quality (SON, 2007). (3) The distribution and sources of heavy metals in the surface water and groundwater are impacted not only by geogenic factors, but also by anthropogenic activities mainly salt mining around a salt pond.

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