



# **AN ASSESSMENT OF HEAVY METALS CONCENTRATION IN WATER AROUND QUARRIES AND BARITE MINE SITES IN PART OF CENTRAL CROSS RIVER STATE, SOUTHEASTERN NIGERIA.**

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## **ABSTRACT**

The assessment of the level of heavy metal concentrations in water sources around quarries and barite mine sites in central Cross River State, Southeastern Nigeria, was carried out to evaluate the degree of contamination due to mining and quarrying activity in the area. The investigation involved the collection of twelve water samples from ponds, streams, river and boreholes around the area. The water samples were passed through 0.45 µm filter paper and preserved with a drop of 0.1M HNO<sub>3</sub> acid. The preserved samples were analyzed for their heavy metal (Ba, Cr, Cu, Fe, Mn, Pb and Zn) content, using inductively coupled plasma-mass spectrometry (ICP-MS). The results show that the average concentration of Ba, Cu, MN, Pb and Zn were above WHO guideline value for drinking water quality, hence not suitable for drinking. The average concentration of the metals is in the order Ba>Mn>Cu>Fe>Pb>Zn>Cr. Computed values of contamination index show that the samples from four locations were contaminated. Single factor pollution index computation indicates that the waters are not contaminated with Cr, but slightly polluted with Fe and Zn, moderately polluted with Ba and severely polluted with Cu, Mn and Pb. Further, Nemerow comprehensive pollution index indicates that samples from four locations were severely contaminated. The study revealed that the water sources around the barite mines had elevated level of heavy metal concentration compared to location around the quarries. It is recommended therefore, that the water be treated before use for drinking purpose.

**KEYWORDS**– heavy metals, contamination index, pollution index, barite mine, quarry.

## **INTRODUCTION**

The central part of Cross River State in Nigeria is very rich in solid minerals, this has led to the proliferation of mining activity in the area by both organize and illegal miners. This surge is attributed to the Federal Government of Nigeria policy on the use of local raw materials. The occurrence and exploitation of these minerals is a potential threat to the water resources within the vicinity of the mines and adjoining area. Rock quarrying and barite mining generates huge quantity of rock and mine tailings. These mine wastes can increase the levels of heavy metals released into the environment. Oxidation and released metals from unprotected mine waste sites are sources of environmental contamination and risk (Adamu *et al.*, 2014). In recent time's heavy metals in soil and water is becoming an issue of concern at all levels, since these resources constitute a crucial component of our environment (Ochelebe *et al.*, 2017).

Mining and release of heavy metals into the environment is a threat to human health because of its toxicity when their levels in drinking water exceeds the maximum admissible values set by international organizations such as World Health Organization. Elevated concentration of elements as a result of contamination can be measured in a number of ways. Hitherto, previous studies have employed the use of indices such as contamination index(CI) and pollution index(PI) (Yang *et al.*, 2013; Adamu *et al.*, 2014; Zhang *et al.*, 2018). Inhabitants within and around the study area mostly depend on water from existing rivers, streams and ponds for domestic purpose. To say the least, few people have access to well-designed and constructed boreholes provided by private owners and Cross River Basin Development Authority, an Agency of Federal Ministry of Water resources through their intervention programs. But this is not enough, so majority still depends on the not too-safe surface water sources such as streams, ponds and river for domestic use. The

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Nigerian National Statistics Office reports that in 2001, 66.5 % of Cross Riverians rely on water from ponds/streams for domestic purposes (FOS, 2003). The Multiple Indicator Cluster Survey report of 2011 shows that 49.3% of Cross Riverians used unimproved water sources for domestic purposes (NBS, 2013). Furthermore, findings of National Nutrition and Health Survey 2014, shows that 37.8 % of household in Cross River State have access to improved drinking water sources (NBS, 2014), indicating that 62.2 % of the population are exposed to unsafe water sources.

Geological, mineralogical and structural aspects of the rocks and barite occurrence have been investigated by previous authors (Ekwueme&Onyeagocha,1986; Ekweme, 1995; Adamu, 2000, 2011; Egeh, *et al.*, 2004; Oden, 2012). Adamuet *al.* (2014),assessed the contamination and health risk associated with abandoned barite mines in Cross River State generally, using streams and pond water.The hydrochemical assessment of pond and stream waters and trace and major element pollution of the sediments around some of the barite mineshave been investigated (Adamuet *al.*,2013; Adamuet *al.*, 2015). However, this study is on the environmental aspect, with emphasis on the effect of the quarry activities and barite mining on available water sources and the risk on human health. The objective is to appraise and update the level of heavy metal in both surface and groundwater sources within and around the quarries and mine sites and appraising the degree of contamination.

#### DESCRIPTION OF STUDY AREA

Six (6) communities namely;Obung, Ojor, Iwuru, Nkonemba, Ikot Okpora and Ibogo in Cross River State, Southeastern Nigeria, where mining is taking place were investigated. During sampling large scale quarrying of granite and granodiorite was going on in Obung, Ojor, Iwuru, Old Netim, Nkonemba and Ikot Okpora area, while barite mining on medium to small scale was going on in Ibogo area. Information from the community leaders revealed thatmodern quarrying activity in the area started around 1971 in Old Netim during the

construction of the major highway in the area, and the latest is the Iwuru quarry that was established around 2012. While barite mining started around 2004 in Ibogo area.

The area lies within longitudes 8°00' to 8°30' E and latitudes 5°20'N to 5°45'N. It is accessed through a major road, the Calabar-Ikom highwayandminor roads such as; Unyanga-lfunkpa road, Ayaba-Ikot Okpora road, Abini-Agwuagune roads, and various footpaths leading to sampling points. The area is drained majorly by Cross River and other smaller rivers and streams(Fig.1). Some of these streams are; Ikpaya, Ayiboniong, Eyuma, Ageden, Ekpendu-Iwuru, Efajene and Ugbam streams. Most of the streams flow in the NE-SW direction. The relief of the area is undulating, with some minor hills and valleys.The mean annual rain fall in the area was reported to be about 2,300mm (CRBDA, 2008). With temperature ranging between 25°C and 35°C (Iloeje, 1991).

#### GEOLOGY

The study area is underlain by two major rock formations, the Oban Massif to the south and the Ikom-Mamfe Embayment to the north. The rocks of the southern part are mainly of igneous and metamorphic origin (granodiorites, phyllites, schists,gneisses, amphibolites, pegmatites, granites, tonalities, monazites, dolerites, and charnockites). The gneisses grade into schist which is intruded by granodiorite and pegmatitein some parts. A sharp contact exists between the schist and calcareous sandstone in the north-western partof the study area (Fig.1). Geochemical studies of the gneisses by Ekwueme and Onyeagocha (1986) shows that they are metasediments of shale-greywacke. The granodiorite is the most extensive intrusive in the study area. The rocks are coarse-grained, non-foliated and have a sharp contact with the schist. Geochemical studies of the schists at Ikot-Ana show them to consist typically ofmetasediments, which have a composition characteristic of phyllites and semi-phyllites. The rock sequence of the Mamfe Embayment consists of sandstones and mudstones(Ekwueme, 1995).

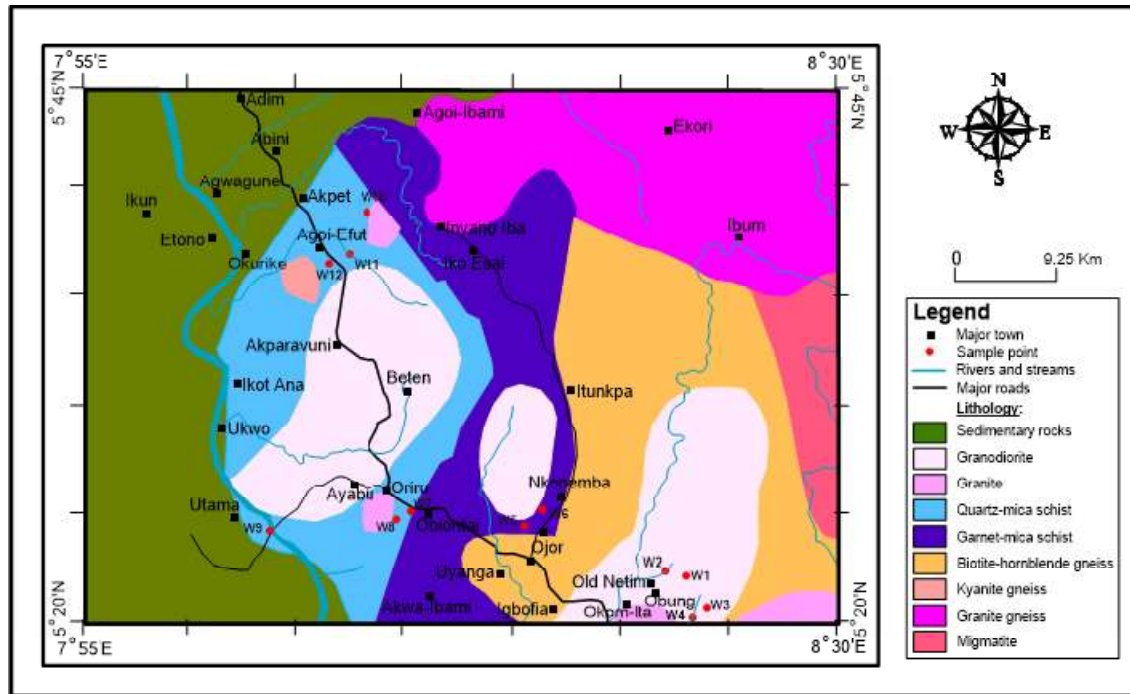


Fig. 1: Geologic Map of the Study Area with sample locations (Modified from Ekwuemeet *et al.*, 1995)

**METHODOLOGY**

A total of twelve (12) water samples consisting of four boreholes, four streams, three ponds and one river, were collected within and around locations of the barites mines and quarries sites (Fig. 1). Table 1 shows the locations and co-ordinates of the sampling points. Water samples were taken from mine ponds, streams and boreholes, and stored in 1 litre polythene bottles. Prior to the collection of water samples, the sample bottles were rinsed thoroughly with the water to be collected. During the collection of groundwater, the borehole was pumped for about 10 minutes to ensure stability, the water samples were then collected at the well head. The

samples were pass through 0.45-  $\mu$ m membrane filter paper.

The samples were preserved with a drop of 0.1M HNO<sub>3</sub>, to prevent precipitation of the metals from the water. The water samples were then stored in a cooler at 4°C and taken to the laboratory for the analysis of their trace element contents using inductively coupled plasma-mass spectrometry (ICP-MS) at Acme Laboratory Limited, Vancouver Canada.

Contamination index was computed to identify the enrichment of heavy metals with respect to the maximum admissible limit (MAL) standards (WHO, 2008). The contamination indexes for

Table 1: Location and Co-ordinates of sampling points.

Loc. ID	Area	Quarry type	Water Type	Co-ordinate	
				Longitude	Latitude
W1	Old Netim	Granite	Pond	E 008.37964°	N 05.36329°
W8	Iwuru	Granodiorite	Pond	E 008.15846°	N 05.42472°
W10	Igbogo	Barite mine	Pond	E 008.14136°	N 05.62878°
W2	Old Netim	Granite	Stream	E 008.37278°	N 05.37422°
W4	Obung	Granite	Stream	E 008.38682°	N 05.34129°
W6	Nkonemba	Granite	Stream	E 008.28652°	N 05.43504°
W11	Igbogo	Barite mine	Stream	E 008.12517°	N 05.60970°
W9	Ikot Okpora	Granodiorite	River	E 008.07167°	N 05.40575°
W3	Obung	Granite	Borehole	E 008.39580°	N 05.34685°
W5	Ojor	Granite	Borehole	E 008.27431°	N 05.40929°
W7	Iwuru	Granodiorite	Borehole	E 008.17486°	N 05.41894°
W12	Igbogo	Barite mine	Borehole	E 008.12000°	N 05.60372°

\*W1-W9= water samples from quarry areas, W10-W12= water samples from barite mine area.

the potentially toxic elements (PTEs) in the water was calculated using Equation. (1). The contamination index is classified as presented in Table 2.

$$C_a = \frac{1}{n} \sum_{i=1}^n \left( \frac{M_{ai}}{S_{ai}} \right) \dots\dots 1$$

Where;

- $C_a$  is the contamination index of sample  $a$
- $M_{ai}$  is the concentration of metal  $i$  in sample  $a$
- $S_{ai}$  is the maximum admissible limit of metal  $i$  base on standard
- $n$  is the total number of metals considered

In order to determine the degree of contribution of each metal to toxicity of the water in the area, the single factor index, Nemerow pollution index (Yang *et al.*, 2013) was useful. The single-factor pollution index can weigh the pollution of single contamination and is applied to establish water quality parameters. The Nemerow comprehensive pollution index is widely applied to reflect the total pollution level and evaluate environmental quality (Zhang *et al.*, 2018). The Nemerow pollution index is calculated using the following equations;

$$F_{ij} = \frac{C_{ij}}{S_{ij}} \dots\dots\dots 2$$

$$PI = \sqrt{\frac{1}{2}(F_{max}^2 + F_{ave}^2)} \dots\dots\dots 3$$

Where;

- $F_{ij}$  is the pollution index of the  $i$ th potentially toxic element in  $j$
  - $C_{ij}$  is the concentration of the  $i$ th potentially toxic element in  $j$
  - $S_{ij}$  is the required standard of the  $i$ th potentially toxic element in  $j$
  - PI is the Nemerow comprehensive pollution index
  - $F_{max}$  is the maximum Single factor pollution index of all the potentially toxic element
  - $F_{ave}$  is the average single factor pollution index of all the potentially toxic element
- the Nemerow pollution index is categorized as presented in Table 2.

**Table 2:** Classes of Contamination and Nemerow Pollution Indices

Contamination Index		Nemerow Pollution Index	
Indices	Degree of contamination	Indices	Degree of contamination
CI < 1	Not contaminated	PI ≤ 1	Not contaminated
1 < CI < 5	Slightly contaminated	1 < PI ≤ 2	Slightly contaminated
CI > 5	Contaminated	2 < PI ≤ 3	Moderately contaminated
		PI > 3	Severely contaminated

**RESULTS AND DISCUSSION**

The result of the heavy metals concentration in the water samples collected and analyzed is presented in Table 3. The result shows that the pH ranged between 5.800 (W11) and 7.200 (W5 and W6), with a mean value of 6.700. The concentration of Ba ranged between 0.010 mg/l (W2) and 13.800 mg/l (W10) with a mean concentration of 1.990 mg/l. The concentration of Cr was below detection limit at locations W2, W3, W4 and W8, with the highest value of 0.030 mg/l (W12). The mean concentration of Cr computed was 0.005 mg/l. Also, Cu was below detection limit at locations W1, W2, W3, W7 and W8, while the highest value was 5.150 mg/l (W10), and a mean concentration of 0.772 mg/l. Fe was below detection at locations W3, W2, W5, W7 and W10 and the highest value of 4.100 mg/l (W10). The mean value computed was 0.586 mg/l. The concentration of Mn was below detection at locations W1, W4, W5, W6, W8 and W9, while the highest value of 4.790 mg/l (W11) was recorded. The mean concentration of 1.110 mg/l

was computed. Concentration values of Pb were below detection limit at locations W1, W2, W3, W4, W5, W6, W7, W8 and W9, while the highest value of 1.130 mg/l (W11) was recorded. The mean concentration value of 0.274 mg/l was obtained for Pb. The concentration of Zn was below detection at locations W1, W5, W8 and W9, and the highest value of 0.270 mg/l (W10) was obtained. However, a mean concentration of 0.079 mg/l was computed for Zn. Generally, the mean concentration of the heavy metals in the area is in the order Ba > Mn > Cu > Fe > Pb > Zn > Cr. Comparison of these data within the different water sources reveals that the concentration of all the metals, except Cr and Zn, were higher in water sampled from the ponds. This conforms with the assertion by Adamuet *al.* (2014). Also, the concentration of all the heavy metals were higher in the samples collected around the barite mines as compared with those of the quarry sites.

**Table 3:** Results of heavy metal concentrations in water samples from the study area.

Loc.ID	Water Type	Ph	Ba (mg/l)	Cr (mg/l)	Cu (mg/l)	Fe (mg/l)	Mn (mg/l)	Pb (mg/l)	Zn (mg/l)
W1	Pond quarry	6.800	0.020	0.003	ND	0.018	ND	ND	ND
W8	Pond quarry	6.700	0.020	ND	ND	0.013	ND	ND	ND
W10	Pond barite	6.700	13.800	0.010	5.150	4.100	4.310	1.104	0.270
<b>Mean</b>		<b>6.733</b>	<b>4.613</b>	<b>0.004</b>	<b>1.717</b>	<b>1.377</b>	<b>1.437</b>	<b>0.368</b>	<b>0.09</b>
W2	Stream quarry	6.600	0.010	ND	ND	ND	0.014	ND	0.002
W4	Stream quarry	7.200	0.020	ND	0.001	0.021	ND	ND	0.002
W6	Stream quarry	6.800	0.020	0.001	0.001	0.039	ND	ND	0.009
W9	River quarry	7.000	0.020	0.001	0.001	ND	ND	ND	ND
W11	Stream barite	5.800	5.600	0.010	1.760	1.800	4.790	1.130	0.210
<b>Mean</b>		<b>6.680</b>	<b>1.134</b>	<b>0.002</b>	<b>0.353</b>	<b>0.372</b>	<b>0.983</b>	<b>0.226</b>	<b>0.045</b>
W3	Borehole quarry	7.000	0.030	ND	ND	ND	0.011	ND	0.003
W5	Borehole quarry	7.200	0.070	0.004	0.001	ND	ND	ND	ND
W7	Borehole quarry	6.700	0.200	0.002	ND	ND	0.021	ND	0.230
W12	Borehole barite	5.900	4.120	0.030	2.350	0.800	4.190	1.056	0.230
<b>Mean</b>		<b>6.700</b>	<b>1.105</b>	<b>0.009</b>	<b>0.588</b>	<b>0.200</b>	<b>1.056</b>	<b>0.264</b>	<b>0.116</b>
<b>Summary of result</b>									
<b>Average around Quarries</b>		<b>6.89</b>	<b>0.046</b>	<b>0.001</b>	<b>0.001</b>	<b>0.010</b>	<b>0.005</b>	<b>0.000</b>	<b>0.027</b>
<b>Average around barite mines</b>		<b>6.167</b>	<b>11.173</b>	<b>0.017</b>	<b>3.087</b>	<b>2.233</b>	<b>4.430</b>	<b>1.067</b>	<b>0.237</b>
<b>Overall Mean</b>		<b>6.700</b>	<b>1.994</b>	<b>0.005</b>	<b>0.7720</b>	<b>0.566</b>	<b>1.111</b>	<b>0.274</b>	<b>0.079</b>
<b>Minimum</b>		<b>5.800</b>	<b>0.010</b>	<b>0</b>	<b>0</b>	<b>0</b>	<b>0</b>	<b>0</b>	<b>0</b>
<b>Maximum</b>		<b>7.200</b>	<b>13.800</b>	<b>0.030</b>	<b>5.150</b>	<b>4.100</b>	<b>4.790</b>	<b>1.130</b>	<b>0.270</b>
<b>WHO</b>		<b>6.5-8.0</b>	<b>0.7</b>	<b>0.050</b>	<b>0.050</b>	<b>0.300</b>	<b>0.300</b>	<b>0.010</b>	<b>0.050</b>

• ND – Not detected

### CONTAMINATION ASSESSMENT

The mean concentration levels of the various parameters analyzed, when compared with standard guidelines for drinking water quality by WHO (2008), show that mean concentrations of Ba, Cu, Mn and Pb obtained in all the water sources were higher (Table 3). It further reveals that concentration levels of Cr are higher in the boreholes, Fe is higher in streams, rivers and ponds. While Zn is higher in ponds and boreholes water.

The results show that the mean concentration of the metals around the quarry sites (W1 to W9) for all the water sources were found to lie within permissible limits for drinking water quality by WHO standard. This indicates that there is no noticeable effect of quarry activities on water quality at present. The locations around the barite mines (W10, W11 and W12), have concentration level higher than the permissible limits for drinking water quality. Water samples obtained and analyzed from the ponds had a higher concentration of heavy metals when compared to those from the boreholes and other surface water. This suggests that activities within and around the barite mines may have adversely affected water quality in the area. The consumption of water with concentration of these heavy metals above the permissible limit could lead to a wide

range of health challenges such as gastrointestinal disorder, neurological disorder, defect in infant mental development and cancer (SON, 2015). Though no information on such health challenges were reported or documented in existing literatures as at the time of this study. However, communities within and around the study area may be at severe health risk by drinking from these water sources.

The contamination indices were computed and the contamination class for each location is presented in Table 4. Comparatively, values of CI >5 indicates high contamination, and these were obtained from locations W10, W11 and W12. These are related to anthropogenic activities and probably resulting from barite mining in the area. Location W7 which is outside the barite mines, had a CI value of 5.0 which indicate contamination, this could be as a result of the high concentration of Zn which contributed to the high index value (Table 5). Therefore, about 33% of the water samples analyzed from the entire study area were contaminated. Other locations such as; W1, W2, W3, W4, W5, W6, W8 and W9, have low average values indicating that they are not polluted with respect to the heavy metals.

Single factor pollution indices (F) and the Nemerow comprehensive pollution Indices (PI) are summarized in Table 5. The average F-values computed were ranked

in the order as follows; Pb>Cu>Mn>Ba>Fe>Zn>Cr. It suggests that Pb, Cu and Mn contributes significantly to

the contamination of the water in the area. The PI values however, indicates that locations W7,

**Table 4:**Results of Contamination Index of the heavy metals in the area

Location ID/ water type	Contamination index	Remark
W1 /Pond quarry	0.150	Not contaminated
W8/Pond quarry	0.070	Not contaminated
<b>W10/ Pond barite</b>	<b>266.750</b>	<b>Contaminated</b>
W2/ Stream quarry	0.100	Not contaminated
W4/ Stream quarry	0.160	Not contaminated
W6/ Stream quarry	0.380	Not contaminated
W9/ river quarry	0.070	Not contaminated
<b>W11/ Stream barite</b>	<b>182.570</b>	<b>Contaminated</b>
W3/ Borehole quarry	0.140	Not contaminated
W5/ Borehole quarry	0.200	Not contaminated
W7/ Borehole quarry	5.000	<b>Contaminated</b>
<b>W12/ Borehole barite</b>	<b>180.320</b>	<b>Contaminated</b>

W10, W11 and W12 are severely contaminated while W1, W2, W3, W4, W5, W6, W8 and W9 are not contaminated. Again, the water sources around the barite mine areas showed high level of contamination compared to those around the quarry areas. This is an indication that barite mining has impacted the area more compared to quarrying.

**Table 5:**Results of single factor pollution index of metals and Nemerow pollution index of the water

Loc. ID/ water type	Ba	Cr	Cu	Fe	Mn	Pb	Zn	F <sub>ave</sub>	F <sub>max</sub>	PI	Remark
W1/ pond quarry	0.030	0.060	0.000	0.060	0.000	0.000	0.000	0.021	0.060	<b>0.045</b>	Not contaminated
W8/ pond quarry	0.030	0.000	0.000	0.040	0.000	0.000	0.000	0.010	0.043	<b>0.031</b>	Not contaminated
<b>W10/ pond barite</b>	<b>19.710</b>	<b>0.200</b>	<b>103.000</b>	<b>13.670</b>	<b>14.370</b>	<b>110.400</b>	<b>5.400</b>	<b>38.117</b>	<b>110.400</b>	<b>82.587</b>	<b>Severely contaminated</b>
W2/ stream quarry	0.010	0.000	0.000	0.000	0.050	0.000	0.040	0.014	0.046	<b>0.034</b>	Not contaminated
W4/ stream quarry	0.030	0.000	0.020	0.070	0.000	0.000	0.040	0.023	0.070	<b>0.052</b>	Not contaminated
W6/ stream quarry	0.030	0.020	0.020	0.130	0.000	0.000	0.180	0.054	0.180	<b>0.133</b>	Not contaminated
W9/ river quarry	0.030	0.020	0.020	0.000	0.000	0.000	0.000	0.010	0.029	<b>0.022</b>	Not contaminated
<b>W11/ stream barite</b>	<b>8.000</b>	<b>0.200</b>	<b>35.200</b>	<b>6.000</b>	<b>15.970</b>	<b>113.000</b>	<b>4.200</b>	<b>26.081</b>	<b>113.000</b>	<b>82.004</b>	<b>Severely contaminated</b>
W3/ borehole quarry	0.040	0.000	0.000	0.000	0.040	0.000	0.060	0.020	0.060	<b>0.045</b>	Not contaminated
W7/ borehole quarry	0.290	0.040	0.000	0.000	0.070	0.000	4.600	0.714	4.600	<b>3.292</b>	<b>Severely contaminated</b>
W5/ borehole quarry	0.100	0.080	0.020	0.000	0.000	0.000	0.000	0.029	0.100	<b>0.074</b>	Not contaminated
<b>W12/borehole barite</b>	<b>5.890</b>	<b>0.600</b>	<b>47.000</b>	<b>2.670</b>	<b>13.970</b>	<b>105.600</b>	<b>4.600</b>	<b>25.760</b>	<b>105.600</b>	<b>76.860</b>	<b>Severely contaminated</b>
<b>Min</b>	<b>0.010</b>	<b>0.000</b>	<b>0.000</b>	<b>0.000</b>	<b>15.970</b>	<b>0.000</b>	<b>0.000</b>	<b>0.010</b>	<b>0.029</b>	<b>0.022</b>	-
<b>Max</b>	<b>19.710</b>	<b>0.600</b>	<b>103.000</b>	<b>13.670</b>	<b>0.000</b>	<b>113.000</b>	<b>5.400</b>	<b>38.117</b>	<b>113.000</b>	<b>82.587</b>	-
<b>Mean</b>	<b>2.850</b>	<b>0.100</b>	<b>15.440</b>	<b>1.890</b>	<b>3.700</b>	<b>27.420</b>	<b>1.590</b>	<b>8.257</b>	<b>27.849</b>	<b>20.436</b>	-

**CONCLUSION**

The investigation reveals that, the mean concentration of Ba, Cu, Mn, Pb and Zn in the water sources within the study area were above the recommended standard for drinking water quality, hence it is not suitable for human consumption. The results of the Contamination Index showed that 33% of the samples were contaminated. The average single factor pollution index, F indicated that the waters are not contaminated with Cr, but slightly contaminated with Fe and Zn, moderately contaminated with Ba and severely contaminated with Cu, Mn and Pb. Furthermore, the Nemerow comprehensive pollution index, PI suggest that 67% of the water samples analyzed are not contaminated, while the remaining samples were severely contaminated. Generally, the areas around the barite mines showed elevated levels of heavy metal concentration compared to locations around the quarries, and the source of these metals may be attributed to the activities of barite mining in the area. It is recommended that water from surface and groundwater sources should be treated before drinking, to forestall health challenges associated with the consumption of water contaminated with these heavy metals.

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