

Original Research

Levels of Lead and Arsenic in Groundwater and Blood of Residents of Agulu, Nigeria

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Abstract

Arsenic and lead come in the first and second positions, respectively, in the Comprehensive Environmental Response, Compensation, and Liability Act (CERLA) list of 275 most hazardous substances of 2011. These metals are capable of bioaccumulation. Therefore, ingestion through food and water should be guarded against. Inhabitants of Agulu, Nigeria, depend mostly on groundwater (wells and boreholes) for their water sources, which exposes them to possible ingestion of these metals through groundwater.

The determination of the heavy metals (HM) arsenic and lead in the groundwaters of Agulu, Nigeria, and their bioaccumulation in the inhabitants was carried out using atomic absorption spectrophotometry. The contamination by these HMs of the sampled groundwater varied among Agulu villages. Arsenic was found to be in more than 41% of the samples, while 40% of them contained arsenic above the 0.01 mg/L MCL recommended by WHO. Lead was detected in 32% of the samples, with 29% being above the WHO MCL of 0.01 mg/L. While the mean blood levels of lead were within allowable limits, the mean blood arsenic levels were several times higher than the allowable limit and far above the water concentrations, which indicated bioaccumulation and chronic arsenic poisoning.

This study has shown significant heavy metal contamination in groundwaters of Agulu and concentrations of arsenic above the allowable threshold. This indicates bioaccumulation of arsenic as the average blood concentration is more than that in the groundwater studied.

Keywords: assessment, heavy metals, exposure, pollutants, groundwater, bioaccumulation

Introduction

The toxic effects of heavy metals on human health are mostly due to exposure to arsenic, lead, mercury, and cadmium. These heavy metals come in the first, second, third, and seventh positions, respectively, in CERLA's list of the 275 most hazardous substances of 2011 [1]. Exposure to heavy metal poisoning is increasing in developing countries such as Nigeria, while exposure has declined in most devel-

oped countries over the last 100 years [2]. Heavy metals are poorly eliminated and can accumulate in the body through many means [3].

Exposure to lead from air and food is roughly equal in proportion to the general population [4]. Exposure to arsenic is mainly via intake of food and drinking water, food being the most important source in most populations [5]. One of the problems associated with low excretion of heavy metals is the potential for bioaccumulation biomagnification, which leads to heavier exposure for some organisms than are present in the environment alone [6].

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Bioaccumulation means an increase in the concentration of a chemical in a biological organism over time, compared to the chemical's concentration in the environment, while biomagnification (also known as bioamplification) is the increase in concentration of a substance that occurs in a food chain as a consequence of persistence (cannot be broken down by environmental processes) and low or non-existent rate of internal degradation/excretion of the substance often due to water-insolubility [7]. The sources of heavy metals that contaminate drinking water supply include industrial and consumer waste and the release of heavy metals into streams, lakes, rivers, and groundwater as a result of acidic rain breaking down soil [8].

In general, a strong relationship between contaminated drinking water with heavy metals and the incidence of chronic diseases such as heart diseases, stroke, cancer, renal failure, liver cirrhosis, hair loss, and chronic anaemia has been documented [9]. Although studies reporting heavy metal pollution in Nigeria abound, there is still no report on the blood levels of arsenic and lead in any section of the population. In this study, the contamination of groundwater by lead and arsenic and the bioaccumulation of these HMs in human blood in Agulu are presented. Arsenic and lead were selected for study because of their high standing in CERLA's list of hazardous substances.

Methods

Our study was carried out in Agulu, which is the most populous town in Anambra state in southeastern Nigeria. Agulu consists of 20 villages. A total of 63 water samples and 62 blood samples were collected and used for analysis (at least three samples were collected from each village). The villages where samples were collected are Umuowelle, Umunnnowu, Umuiifite, Umubiala, Ukunu, Uhueme, Okpu Ifite, Okpu Agulu, Odidama, Obeagu, Obe, Nwanchi, Nnohia, Nneogidi, Nkitaku, Isiamigbo, Ifiteani, Amaoji, Amaezike, and Amaatutu. The geographical location of Agulu is 6°07'N 7°04'E.

The groundwater samples were randomly collected in prewashed (with detergent, doubly de-ionized distilled water, dilute nitric acid and doubly de-ionized distilled water, sequentially) bottles during the month of July 2012. The samples were acidified to 1% with nitric acid (Spectra, South Africa) and subsequently stored at 4°C before analysis within 24 hours of sample collection. The water samples were analyzed for the presence of arsenic and lead using an FS 240 atomic absorption spectrometer (Agilent technologies, USA).

The calibration plot method was used for the analysis using standard solutions of lead (Spectra, South Africa) and arsenic (Spectra, South Africa) respectively. Air-acetylene (for lead) and nitrous oxide-acetylene (for arsenic) were the flame gases used, and hollow cathode lamp of the corresponding elements supplied the resonance line source. The wavelengths for the determination of the elements were 217.0 nm and 193.7 nm for lead and arsenic, respectively. The samples were analyzed in duplicate with the average

concentration of the target analyte displayed in mg/L by the instrument after interpolation from the standard curve of each element.

Blood serum from Agulu residents more than 50 years of age were used for the study. Samples were collected from three different families from each village in Agulu who have inhabited the area for over 50 years.

A minimum of 3 ml and a maximum of 5 ml of venous blood (enough to obtain a good quantity of serum to be used) was drawn (with total consent of the individuals following the operational guidelines laid down by the Nnamdi Azikiwe University Ethics committee for ethical review of research on human subjects) into a sterile vacutainer tube (MDM, China) containing no anticoagulant or contaminant. The collected sample(s) was kept in upright position at room temperature for 30–45 min to allow clotting and was centrifuged for 15 min at 1000–2000 rpm using an 80-2B centrifuge (Ningbo Hinotek technology co., Ltd, China). With the aid of a pipette, the serum (supernatant) was aspirated into a cryovial tube and stored at 4°C until the sample was analyzed.

The samples were each diluted with deionized water at 1:5 dilution so that the sample solution falls within the absorbing range. The dilutions were filtered with the aid of a filter paper (Whatman No. 1) to remove any extraneous materials that could interfere with analysis.

The prepared samples were analyzed for lead and arsenic using FS 240 atomic absorption spectroscopy (Agilent technologies, USA). The analysis was carried out at a wavelength of 217.0 nm and 193.7 nm for lead and arsenic, respectively. Air-acetylene flame (for lead) and nitrous oxide-acetylene flame (for arsenic) were used. The resonance sources were hollow cathode lamps of the corresponding element (lead and arsenic hollow cathode lamp). The automated Atomic Absorption Spectrometer applied the calibration plot method and this was used to obtain the concentrations (displayed in mg/L) of arsenic and lead, which were interpolated from the standard calibration curve.

The mean concentrations of the individual metals in both the groundwater and blood per village were subjected to a statistical test to verify the correlation between the means and any significant difference. Ethical approval of this part of the study involving human beings was obtained from Nnamdi Azikiwe University Ethics committee for ethical review of research on human subjects.

Results and Discussion

In the analysis of the water samples (63) collected in Agulu for lead, 43 samples (68.25%) did not contain detectable levels of lead, while 20 samples (31.75%) contained lead. The lead contents ranged from 0.01 mg/L – 0.51 mg/L. Of the 20 samples containing lead (13 well water and seven boreholes), 18 (28.57%) of them were in concentrations above the maximum contaminant level (MCL) of 0.01 mg/L.

A total of five villages did not contain detectable levels of lead (Isiamigbo, Uhueme, Umubiala, Umunnnowu, and

Table 1. Mean concentrations of lead and arsenic in samples from different villages in Agulu.

No.	Village	Blood sample		Water samples	
		Lead (mg/l)	Arsenic (mg/l)	Lead (mg/l)	Arsenic (mg/l)
1	Amaatutu	0.35	5.83	0.147	0.027
2	Amaezike	0.32	13.60	0.095	0.103
3	Amaoji	0.08	0.00	0.093	0.133
4	Ifiteani	0.11	3.56	0.07	0.1
5	Isiammaigbo	0.33	0.00	0	0.003
6	Nneogidi	0.31	2.46	0.17	0.017
7	Nneoha	0.33	14.43	0.023	0.083
8	Nnkitako	0.10	0.22	0.077	0.177
9	Nwanchi	0.17	14.37	0.057	0.057
10	Obeagu	0.33	9.17	0.083	0.103
11	Obe	0.22	0.00	0.117	0.037
12	Odidama	0.05	0.83	0.117	0.15
13	Okpu Agulu	0.43	7.40	0.083	0.077
14	Okpu Ifite	0.42	9.55	0.127	0.137
15	Uheme	0.28	0.00	0	0.043
16	Ukunu	0.18	0.00	0.03	0.06
17	Umubiala	0.23	7.81	0	0.007
18	Umuifite	0.08	3.50	0.01	0.027
19	Umunnonwu	0.07	6.90	0	0.017
20	Umuowelle	0.05	2.00	0	0

Umuowelle) from any of their three water samples. Nkitaku had the highest concentration of lead, with average concentration of 0.17 mg/L, while Umuifite, with average lead concentration of 0.01 mg/L, had the least.

The concentrations of arsenic in the analyzed water samples varied from 0.01 mg/L – 0.45 mg/L. On the other hand, a total of 37 samples (58.73%) did not contain detectable levels of arsenic, while 26 samples (41.27%) spread across all the villages except Umuowelle contained arsenic. Neither arsenic nor lead was detected in any of the three samples from Umuowelle. Of the 26 samples (16 well water and 10 borehole samples) containing arsenic, 25 (39.68%) were in concentrations above the MCL (0.01 mg/L), with the maximum concentration being 0.45 mg/L (Table 1).

The highest and lowest concentrations of arsenic were recorded in Nneohia and Isiammaigbo, respectively, with their respective average arsenic concentrations being 0.177 mg/L and 0.003 mg/L (Table 1).

The differences in heavy metal contamination between villages may be attributed to the different characteristics of the media through which the water passes on its way to the groundwater zone of saturation, agricultural practices, nat-

ural composition of the soils, nearness to industries, car mechanic workshops, or refuse dumps, and proximity to areas with congested traffic and use of cements to plaster the walls of wells [10].

Agulu is not industrialized, although it is about 20 km (as the crow flies) away from Nnewi town, which is heavily industrialized. In a vertical migration of heavy metals in dumpsite soil, downward migration from the reference has been reported. All the heavy metals (Cu, Pb, Mn, and Zn) that have been determined showed higher concentrations toward the dumpsite [11].

Fig. 1 shows a significant correlation between the distance of the villages from Nnewi and the average concentration of heavy metals in the sampled groundwater. Although this is possible, more data may be needed to conclude that migration was solely responsible for the contamination. Arsenic contamination in groundwater is mostly from geological sources. The primary source of contamination is from bedrock and this occurs due to heavy mining activities [12]. However, there is no heavy mining activity close to the area of study; rather, the famous Agulu-Nanka gully erosion sites [13] are located around the study area, although processing of ores in the industrialized Nnewi

area and Awka could constitute anthropogenic sources. The washing away of the upper layers of the earth's crust by erosion would result in exposure of the bedrock, leading to contamination of groundwater by arsenic. Also, in some areas drilled boreholes and wells may pass through the same bedrock that contains arsenic.

The absence of lead in the samples from Umuowelle, Umubiala, Uhueme, Isiamigbo, and Umunnowu can be attributed to the combined effects of distance from the dumpsite and the dilution effect of Agulu lake, which is located in Umuowelle, while Umubiala is within about 1.2 km circumference of the lake. It has been reported that the aquifer around the lake lies between 3 and 30 metres [14].

The dilution effect of the lake, which gives rise to the Idemili River, could explain the absence of heavy metals in the sample from Umuowelle, and the absence of lead from Umubiala. Isiamigbo is 2.5 km from another body of water, indicated in Google maps as a stream known locally as Ududonka. This could account for the absence of lead and the lowest concentration of arsenic in the samples from there. The trend shows that the concentration of the metals tends to diminish the farther the village is located from Nnewi town. The absence of lead in Umunnowu and Uhueme samples could be largely due to the distance from Nnewi. The heavy metal contamination in Agulu is most likely due to vertical migration from Nnewi. This implies that the concentration of the studied heavy metals in groundwaters of Nnewi could be higher. The health implications of the groundwater contamination by these HMs to the inhabitants of Agulu will be bioaccumulation that normally leads to grave health implications that would have gone unnoticed.

The concentration of lead and arsenic in the blood samples collected from the 20 villages were in most cases higher than what was obtainable from the water sources investigated.

The differences between the mean blood concentration of lead and mean water concentration were significant

($p < 0.05$). Also, the differences between blood and water concentrations of arsenic were also significant ($p < 0.05$). This indicated bioaccumulation as the blood concentrations were far above those of the water concentrations. Drinking water is one of the major sources of heavy metal exposure [15]. However, villages such as Isiamigbo, Uhueme, and Umubiala, which had no lead in water samples, had appreciable mean concentrations of lead in blood samples (Table 1). This suggested that sources of lead accumulation are more than water. Other sources of lead accumulation include food and air [16]. The mean blood lead levels, however, were below WHO's 0.045 mg/dl, above which treatment should be initiated. On the contrary, most of the arsenic mean and individual blood levels were far above the acceptable blood level, which is < 0.001 mg/L [17]. These concentrations were several times more than the water concentration of the arsenic in respective villages, indicating serious bioaccumulation over the years. However, Umuowelle village, which had no detectable arsenic in water samples, had up to 2 mg/L mean blood level of arsenic, suggesting that drinking water may not be the only source of arsenic bioaccumulation in the blood samples. Also, average accumulation was more among males (5.45 mg/L) compared to females (4.6 mg/L). This position is corroborated by an earlier report that males are higher accumulators of arsenic than females [7]. Considering that there have been several reported cases of chemical poisoning in Nigeria [18], this study, which shows that there are some cases of unreported chronic chemical poisoning, calls for concern – especially as the symptoms of chronic arsenic poisoning such as headache, diarrhoea, drowsiness, and convulsion are associated with common ailments and are therefore not specific, which could lead to misdiagnosis [19]. Also, arsenic poisoning is associated with heart disease, stroke, and cancer, among others [20]. Several cases of myocardial infarction and arterial thickening in children who consumed water containing about 0.6 mg/l arsenic [21]. Low-level arsenic exposure by humans may also

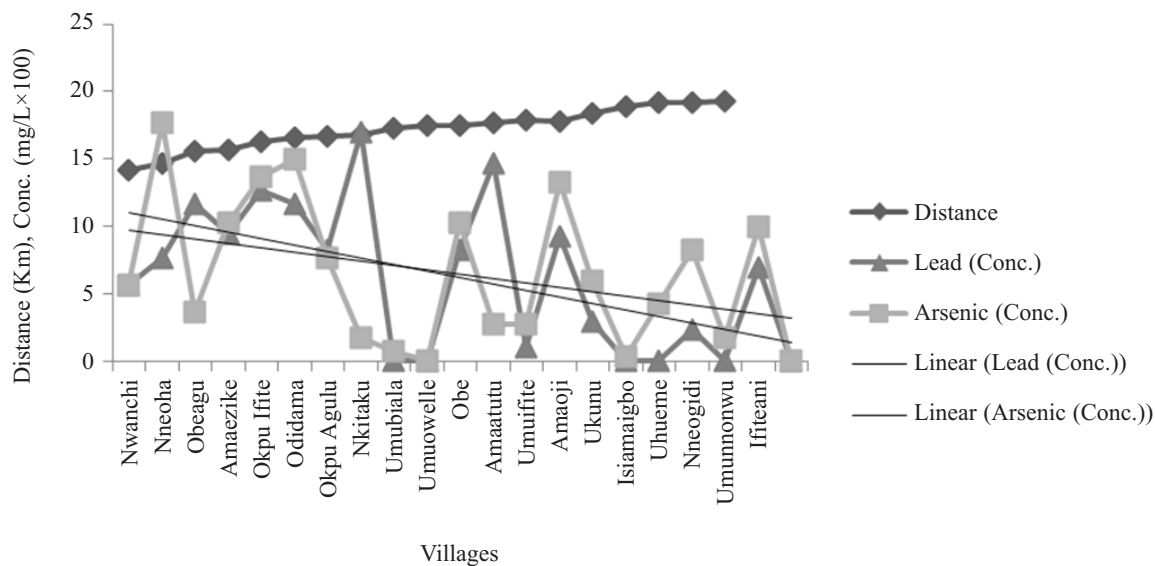


Fig. 1. Correlation between concentrations of heavy metals and the distance of villages from Nnewi, a heavily industrialized town.

cause vascular system damage, a classic example of which is Blackfoot disease, which was endemic in an area of Taiwan where most drinking water contains 0.17 to 0.8 ppm arsenic [22].

These resultant conditions are among the most common causes of death in Agulu and Nigeria, in general contributing about 11% of all deaths and 46% of deaths from non-communicable diseases [23, 24]. Therefore, high blood levels of arsenic have contributed unarguably to the poor health conditions and short life expectancy in an area where approximately 400 out of 1,000 adults die before age 60 [25].

The measurement of groundwater contamination by lead and arsenic in Agulu showed that more than a significant number of the samples contained heavy metals at a concentration above WHO's recommended maximum contaminant level.

Conclusion

This study showed that the groundwater in Agulu is contaminated with significant quantities of the heavy metals (lead and arsenic) analyzed. Measurement of the levels of the lead and arsenic in human blood showed that while the mean blood lead levels were within acceptable limits, the mean blood levels of arsenic were far above the acceptable limits in human blood. Steps should be taken to determine the major source of arsenic exposure as it did not seem to come from groundwater alone.

Abbreviations

CERLA – Comprehensive Environmental Response, Compensation, and Liability Act
 WHO – World Health Organization
 MCL – Maximum contaminant level
 HM – Heavy metal

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