Determination of Radioactivity and Heavy Metals Concentration in Underground Water Sources in Three Selected Local Government Areas of Benue State

Egede Owulo Hope[1], N. B. Akaagerger[2], B. A. Ikyo[3]

[1, 2, 3]Department of Physics, Benue State University Makurdi

egedehope[at]gmail.com, bnguvan[at]gmail.com, iakyo[at]bsumedu. ng,

Abstract: ***The Heavy metals (Cadmium, Manganese, Copper, Nickel and Lead) concentration and activity of radionuclides (gross alpha and beta) from boreholes and surface wells in Oju, Obi and Otukpo Local Government Areas of Benue State. They were determined using Atomic Absorption Spectrometer and Proportional Counter respectively. The results obtained show that the mean concentrations of heavy metals for borehole water samples are; Cd: 0.103 mg/L; Cu: 4.749 mg/L; Mn: 7.313 mg/L; and Ni: 1.376 mg/L; and that for surface wells are; Cd: 0.092 mg/L; Cu: 10.756 mg/L; Mn: 8.011 mg/L; and Ni: 1.238 mg/L. The Pb concentration was below detection limit in both water sources. The mean activity concentration for water samples was found to be: borehole 0.041 ± 0.004 Bq/L and 0.071 ± 0.007 Bq/L; surface well: 0.022 ± 0.002 Bq/L and 0.058 ± 0.003 Bq/L for alpha and beta respectively. The values of the heavy metals concentrations are higher compared to Nigerian Standard on Drinking Water Quality (NSDWQ) (Cd: 0.003 mg/L, Cu: 2 mg/L, Pb: 0.01 mg/L, Mn: 0.4 mg/L and Ni 0.07) and World Health Organization (WHO) (Cd: 0.003 mg/L, Cu: 1 mg/L, Pb: 0.01 mg/L, Mn: 0.2 mg/L and Ni 0.02) except for Pb which was below detection limit. The results revealed that the boreholes and surface wells in the study area have become contaminated by heavy metals discharged into them. However, the measured gross alpha and beta activity were below the reference level set by WHO (0.500 Bq/L for gross alpha and 1.000 Bq/L for gross beta). Similarly, the calculated annual committed effective dose for adults and children were all below the Individual Dose Criterion (IDC) value of 0.1mSvy^{-1}. The low alpha and beta activity concentrations in boreholes and surface wells show that there is low natural radionuclide activity of the underlying rocks in the areas. By this finding radioactivity in the water samples is not a threat to the users in the study area, however heavy metal concentrations is a problem.***

1. Introduction

It is estimated that 2.5% of the global water resources (about 35 million km^3) consists of freshwater. This is merely a fraction of water available for domestic use. Some of the 30% fresh water stored as groundwater is found in rocks, and it is contaminated with heavy metals (Ashton, *et al*., 2012).

The Heavy metals are accumulated into the soft tissues of the body when water from these sources is consumed. Although, the presence of some heavy metals is essential to the proper functioning of the human body, most are poisonous to the human system. Metals essential to the human body include; Cu, Zn, Cr, Fe and Mn. When the recommended doses of these metals are ingested, they become poisonous to the human body. The most common heavy metals that cause serious damages to the human health are Pb, Hg, As and Cd. Heavy metal poisoning can result from the following: industrial exposure, water or air pollution, foods, drugs, improperly coated food containers or the ingestion of lead-based paints (Shittu, Ndikilar and Suleiman 2016). Water is used practically in every vital human tasks and procedures. It is a vital component in both household and additionally mechanical and industrial purposes.

It is found that heavy metals in mining areas and old mine sites, cause environmental pollution, this pollution effect reduces with increasing distance away from mining sites (Peplow, 1999). These metals are leached out along slopes, and are carried by acid water downstream or run-off to the sea. Through mining activities, water bodies are most emphatically polluted (INECAR, 2000).

Water from the ground can be contaminated by the deposition of radioactive materials (WHO, 1998). These radioactive materials include Uranium (^{238}U) and Thorium (^{243}Th), and its progenies, Radon and Thoron. They contaminate rain and groundwater which are sources of drinking water, and as a result, deep wells and boreholes contain high concentrations of radioactive elements. Human activities such as mining, milling and processing of uranium ores and mineral sands, Manufacture of fertilizers, drilling, transportation, agricultural activities, processing and burning of fossil fuels have raised the concentrations of naturally occurring radioactive materials in the environment (Pujol and Sanchez-Cabeza, 2000). Radioactive materials could also be washed into wells, boreholes and even enter through burst pipes. Important radioactive elements in drinking water are tritium, potassium-40, radium and radon which are alpha or beta emitters (Surbeck, 1995).

Intake of contaminated water on humans and animals causes cancers, toxicity of the kidneys or children with birth defects, irregularity in blood composition, adverse effects on vital organs such as kidneys and liver disorder (WHO, 2006). Alpha particles are heavy particles and can only move a short distance. It cannot permeate through our skin but it can be inhaled and settled in the lung tissue. The energy deposition in lungs is highly localized and it is said to elevate the risk of lung cancer (UNSCEAR, 2000).

Toxic elements (metals) are usually present in industrial, municipal and urban runoff, which can be harmful to biotic life. Increased urbanization and industrialization are causes of increased level of trace metals, especially heavy metals, in our waterways. Many dangerous chemical elements if released into the environment accumulate in the soil and sediments in water bodies (Serife, Kartal and Latif *et al*, 2000).

There are over 50 elements that can be classified as heavy metals, of which 17 are considered to be very toxic and relatively accessible. Characteristically, anions play important role in drinking water. Heavy metals have a marked effect on the aquatic flora and fauna, which also through biomagnifications (Chaitali and Jayashree 2013) enters the food chain and ultimately affect humans as well.

Studies report various effects of heavy metals in drinking water (US Environmental Protection Agency (USEPA 2015), Agency for Toxic Substances and Disease Registry (ATSDR 2015). According to the International Agency for Research on Cancer (IARC), inorganic Arsenic (As) and Cadmium (Cd) are classified as human carcinogens (IARC, 2016). As is related to cancer risk and skin damage, Cadmium (Cd) is linked to kidney damage and cancer. Other effects such as heart diseases and blood cholesterol from Sb, Anemia from Pb, kidney and liver damage from Hg, and gastrointestinal disorder from Cu are also reported (USEPA 2015, ATSDR 2015).

Drinking water is obtained from a variety of sources like wells, rivers, lakes, boreholes, reservoirs and ponds. The various sources of water pose the greatest risk to human health due to contamination of these sources. Water pollutants mainly consists of heavy metals, microorganisms, fertilizers and thousands of toxic organic compounds. Generally, radionuclides are also present in water in varying amounts from natural sources within the Earth or due to releases from nuclear power plants or laboratories. Water from wells, for example, can be exposed to toxins from rock formations that can contribute radiological toxins like uranium, radium and thorium. Although all water contains some level of radiation, the type and amount are dependent on a variety of factors. The most common naturally occurring alpha particles in rocks and soil are radium-226, uranium-238, radon-222, polonium-210 and lead-206. The primary beta particles typically are manmade, like strontium-90, but some are naturally occurring, like potassium-40. Some of the decay products from radon also emit beta particles. Higher levels of radiological contaminants can be found in groundwater near mining operations or areas where rock and soil have been disturbed (Marianne Metzger 2014).

2. Study Area

The research focused on measurement of radioactivity and heavy metals concentration in drinking water from three (3) Local Government Areas in Benue South East Zone, Nigeria. The Local Government Areas are: Oju, Obi, and Otukpo. The coverage of this work is limited to selected local government areas which include Oju, Obi, and Otukpo Local Government Areas of Benue State. This investigation is limited to underground water sources (hand surface wells and boreholes) used by people for drinking, domestic activities, irrigation and livestock.

3. Materials and Methods

3.1 Materials

The materials used for this research work include; Atomic Absorption Spectrometer (AAS), MPC 2000DP Counting system, Plastic containers, Hand gloves, Meter ruler, Concentrated Nitric acid, Syringes, Ethanol, Masking tape, Hot Plate, Beakers, Acetone, Graduated measuring cylinders, Ceramic petri-dishes, Planchets, Stirring rod, Spatulas, Detergent, Vinyl acetate, Distilled water, Filter paper, Analytical weighing balance, Glass desiccators and Funnel.

3.2 Method

Heavy metals in water were determined by the use of Atomic Absorption Spectrometer (AAS) in Chemistry Laboratory in Benue State University, Makurdi. About 70 cm^3 volume of each water sample was evaporated on a hot plate at a temperature below the boiling point of water till the volume reduced to 50 cm^3; 5 cm^3 of Concentrated HNO_3 was then added and heated in an evaporated disc to reduce the volume to 20 cm^3. After cooling, 5 cm^3 of Concentrated HNO_3 was added to the sample and heated until a residue was formed. Finally, the residue was washed down with distilled water and then filtered to remove insoluble materials that could clog the atomizer. In each case, the filtrate volume of the sample was made up to70 cm^3 with distilled water for analysis of heavy metals.

3.3 Counting and Analysis

Acetone (a cleansing agent) was first used to wash the apparatus for the sample preparation. About 1000 cm^3 of the sampled water was measured and transferred to a beaker and placed on the hot plate. The sample was evaporated carefully on a temperature adjustable hot plate in order to achieve the required residue. The evaporation was done at the temperature less than 100 ^{0}C until the volume reduced to about 50 cm^3, the sample volume was transferred to an empty weighed petri- dish (g1), the petri-dish with its content from the 50 cm^3 of the sample evaporated was placed on the hot plate or under the infrared radiator lamp for surface evaporation and drying to obtain the required residue. The petri-dish and the dried residue was weighed (g2). The weight of the residue obtained from 1000 cm^3 of the sample evaporated was obtained by subtracting the weight of the empty dish from the weight of the dried residue and the dish (g2- g1). The weight of the residue required for measurement is 0.077 g. The specification by International Organization for Standardization (ISO) is, 0.077 g of the residue was used for analysis. The residue 0.077 g was transferred into sterilized planchet and weigh on the analytical balance. The desired weight of 0.0770 *g* of residue on the planchet was transferred to the sample holder

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of MPC 2000 model detector. The detector was operated in alpha and beta modes to obtain the count rates of alpha and beta in counts per minutes respectively. (Shittu, *et al.*, 2016).

The alpha and beta count rate and Radioactivity were calculated using the following equations

Count Rate (R)

$$R(\alpha, \beta) = \frac{counts\ (\alpha, \beta)}{Elapsed\ minutes} \quad (1)$$

$$\text{Radioactivity (Bq/l)} = \frac{\alpha\ or\ \beta(cpm) - BKGCountrate\ (cpm)}{SE \times SS \times DE} \times \frac{1}{60} \quad (2)$$

Where A is the Radioactivity, SE is the Sample Efficiency, SS is the sample size or equivalent volume of water evaporated, DE is the Detector Efficiency, BKG is the background count rate of the detector.

4. Results

Water samples for boreholes and surface well water were studied. The analysis was done for radioactivity and heavy metals concentration.

4.1 Elemental Analysis

The dominant heavy metals found in this research are: Cadmium (Cd), Copper (Cu), Manganese (Mn), Nickel (Ni), Lead (Pb). A chart of heavy metals of borehole water, and hand surface well water are presented below in Table 1 and 2 respectively.

A chart of heavy metals in borehole water is presented below in figure1.

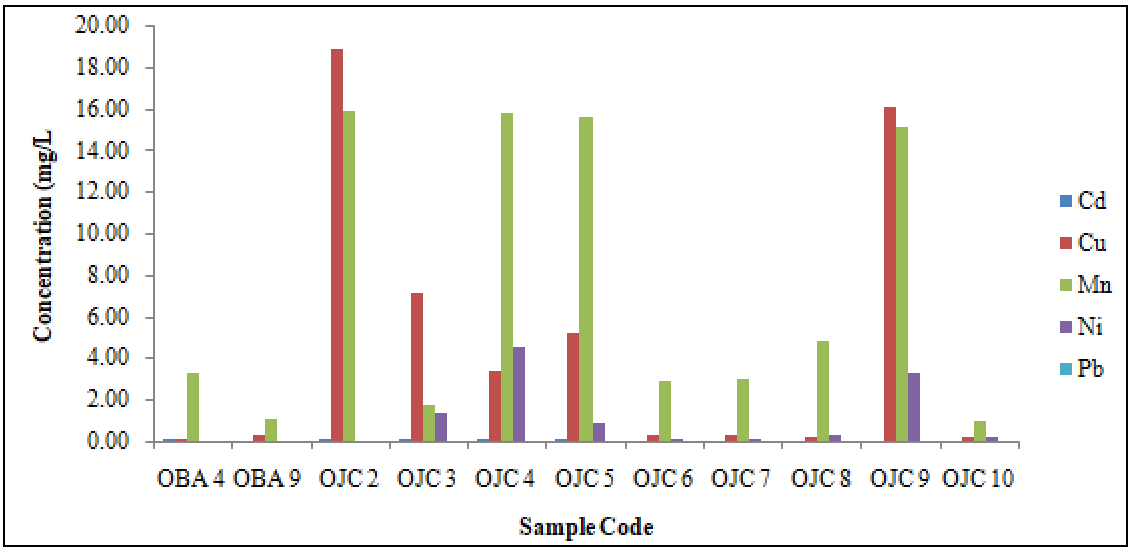

Figure 1: Measured heavy metals concentration of borehole water samples

A chart of heavy metals in hand dug water is presented below in figure 2.

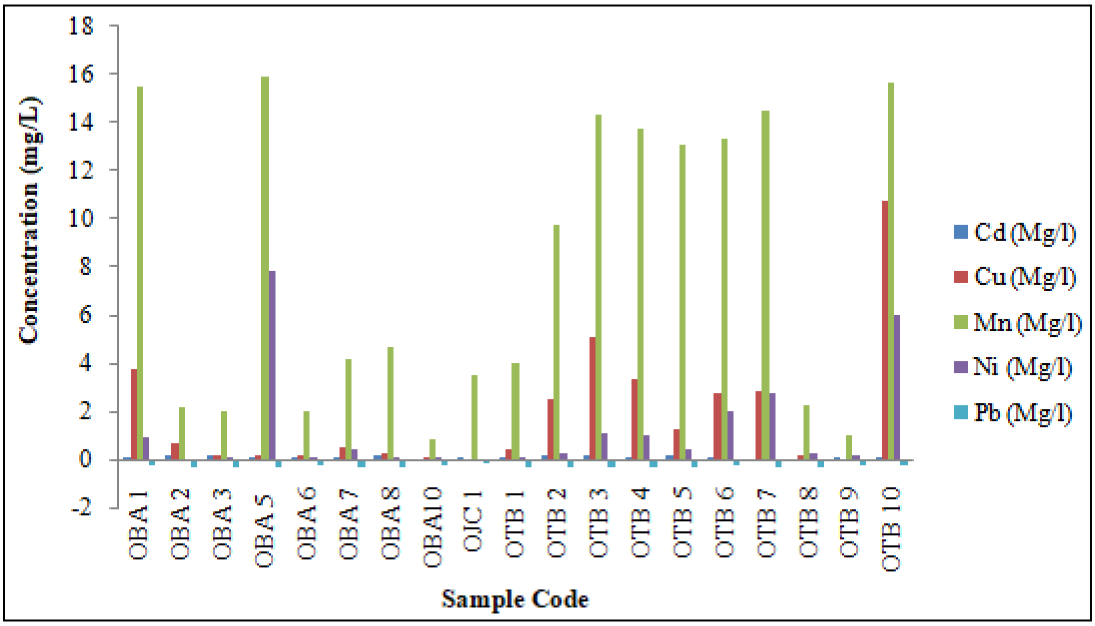

Figure 2: Measured heavy metals concentration of hand surface well water samples

Table 1: Alpha and Beta activity with annual committed equivalent dose for Borehole water

Sample ID	Alpha (α) Activity (Bq/L)	Beta (β) Activity (Bq/L)	α -Annual Committed Effective Dose ($mSvyr^{-1}$)		β -Annual Committed Effective Dose ($mSvyr^{-1}$)	
			Children 1-12yrs	Adult ≥ 12yrs	Children 1-12yrs	Adult ≥ 12yrs
OBA 4	0.022 ± 0.002	0.110 ± 0.005	0.003	0.006	0.014	0.029
OBA 9	0.081 ± 0.003	0.137 ± 0.006	0.011	0.021	0.018	0.036
OJC 2	0.004 ± 0.002	0.021 ± 0.004	0.001	0.001	0.003	0.005
OJC 3	0.024 ± 0.002	0.021 ± 0.003	0.003	0.006	0.003	0.005
OJC 4	0.024 ± 0.004	0.068 ± 0.009	0.003	0.006	0.009	0.018
OJC 5	0.069 ± 0.004	0.094 ± 0.008	0.009	0.018	0.012	0.025
OJC 6	0.025 ± 0.007	0.034 ± 0.013	0.003	0.007	0.004	0.009
OJC 7	0.015 ± 0.004	0.036 ± 0.008	0.002	0.004	0.005	0.009
OJC 8	0.079 ± 0.005	0.174 ± 0.005	0.010	0.021	0.023	0.045
OJC 9	0.041 ± 0.002	0.103 ± 0.005	0.005	0.011	0.013	0.027
OJC 10	0.033 ± 0.004	0.077 ± 0.008	0.004	0.009	0.010	0.020

Table 2: Alpha and Beta activity with annual committed equivalent dose for surface well water.

Sample ID	Alpha (α) Activity (Bq/L)	Beta (β) Activity (Bq/L)	α -Annual Committed Effective Dose ($mSvyr^{-1}$)		β -Annual Committed Effective Dose ($mSvyr^{-1}$)	
			Children 1-12yrs	Adult ≥ 12yrs	Children 1-12yrs	Adult ≥ 12yrs
OBA 1	0.034 ± 0.002	0.086 ± 0.004	0.004	0.009	0.011	0.022
OBA 2	0.010 ± 0.002	0.067 ± 0.004	0.001	0.003	0.009	0.018
OBA 3	0.019 ± 0.001	0.060 ± 0.003	0.002	0.005	0.008	0.016
OBA 5	0.013 ± 0.001	0.038 ± 0.002	0.002	0.003	0.051	0.010
OBA 6	0.020 ± 0.001	0.032 ± 0.002	0.003	0.005	0.004	0.008
OBA 7	0.013 ± 0.001	0.031 ± 0.002	0.002	0.003	0.004	0.008
OBA 8	0.012 ± 0.001	0.051 ± 0.004	0.002	0.003	0.007	0.013
OBA 10	0.028 ± 0.002	0.051 ± 0.003	0.004	0.007	0.007	0.013
OTB 1	0.026 ± 0.002	0.053 ± 0.003	0.003	0.007	0.007	0.014
OTB 2	0.046 ± 0.003	0.094 ± 0.005	0.006	0.012	0.012	0.025
OTB 3	0.054 ± 0.003	0.101 ± 0.005	0.007	0.014	0.001	0.003
OTB 4	0.021 ± 0.001	0.077 ± 0.003	0.003	0.005	0.010	0.020
OTB 5	0.029 ± 0.002	0.054 ± 0.003	0.004	0.008	0.007	0.014
OTB 6	0.038 ± 0.003	0.084 ± 0.005	0.005	0.010	0.011	0.022
OTB 7	0.012 ± 0.001	0.053 ± 0.003	0.002	0.003	0.007	0.014
OTB 8	0.016 ± 0.001	0.048 ± 0.003	0.002	0.004	0.006	0.013
OTB 9	0.030 ± 0.002	0.082 ± 0.004	0.004	0.008	0.011	0.021
OTB 10	0.002 ± 0.001	0.041 ± 0.002	0.000	0.005	0.005	0.011
OJC 1	0.002 ± 0.001	0.007 ± 0.003	0.000	0.005	0.001	0.002

Table 3: The range and the mean values of measured heavy metal for Borehole water

Heavy metals	Range (mg/L)	Mean (mg/L)
Cd	0.040-0.171	0.103
Cu	0.101 – 18.969	4.749
Mn	0.977 – 15.953	7.313
Ni	0.044 – 4.899	1.457
Pb	BDL – BDL	BDL

Table 4: The range and the mean values of measured heavy metal for surface well water

Heavy metals	Range (mg/L)	Mean (mg/L)
Cd	0.027 – 0.168	0.092
Cu	BDL – 10.756	1.685
Mn	0.796 – 15.902	8.011
Ni	0.011 – 7.835	1.238
Pb	BDL – BDL	BDL

Table 5: Mean values of Gross Alpha and Beta Radioactivity and Annual Committed Equivalent Dose (CED).

Sample type	Alpha (α) Activity (Bq/l)	Beta (β) Activity (Bq/l)	ACED (α) ($mSvyr^{-1}$) Children	ACED (α) ($mSvyr^{-1}$) Adult	ACED (β) ($mSvyr^{-1}$) Children	ACED (β) ($mSvyr^{-1}$) Adult
Borehole	0.041	0.071	0.005	0.010	0.010	0.021
Well	0.022	0.058	0.003	0.006	0.007	0.013

5. Discussion

Studies have shown contamination of soil and water samples by metals as a result of agricultural, manufacturing, pharmaceutical, industrial or residential activities (Abolude *et al.*, 2013). In this work; cadmium, copper, manganese, nickel and lead were found in varied concentrations.

The concentration of heavy metals in the sample for borehole water with Cadmium ranging from 0.04-0.171 mg/L and all the values are higher than the reference values set by the WHO (0.003 mg/L) and NSDWQ (0.003 mg/L).

The cadmium concentrations in all the borehole samples are higher than the reference value given by NSDWQ (0.003 mg/L), WHO (0.003 mg/L) and USEPA (0.005 mg/L). Much work has been done on heavy metal concentration in the studied samples. For example, Abolude *et al.*, 2013 studied heavy metals in the sewage pond and found chromium, manganese, iron, copper, nickel, cobalt, zinc, arsenic and lead. In their work they attributed high values of Mn and Cu to the storage batteries, paints and inks in several products like phosphate fertilizers detergents and refined products. In this work, the high values of Mn and Cu could be attributed to the use of phosphate fertilizers, detergents, used batteries and other chemicals like fungicides. Ehi-Eromosele and Okiei in a study showed that ingesting copper for instance may also lead to coronary heart diseases and high blood pressure, although coronary heart diseases have also been linked to copper deficiency Ehi-Eromosele and Okiei, 2012. Copper has its highest concentration in OJC 2 with 18.969 mg/L. Farming activities have been a predominant occupation within the localities where the samples were collected. The high concentration of copper could be attributed to contamination from soil through leaching, the use of fertilizers and fungicides in agricultural practices among the famers within the vicinity where the boreholes were sank.

Copper has its highest concentration in OJC 2 with 18.969 mg/L for borehole water. The concentration of copper in OBA 4, OBA 9, OJC 6, OJC 7, OJC 8 and OJC 10 are lower than the recommended values set by NSDWQ (1.0 mg/L) and (2.0 mg/L) by WHO while OJC 2, OJC 3, OJC 4, OJC 5, and OJC 9 are higher than the reference values of WHO and NSDWQ. The values of copper concentration in surface wells ranges from BDL-10.756 mg/L with a mean of 1.685 mg/L for surface well. All the values are higher than the reference levels set by WHO (2 mg/L) and NSDWQ (1.0 mg/L) except OTB 9 with value BDL. Introduction of some agricultural practices such as application of inorganic manure such as fertilizers and the use of fungicides could be attributed to the high value just as leaching from the soil may not be exempted.

The rampant use of fertilizers and agricultural chemicals such as herbicides as well as leaching from the soil and rocks could be a reasonable factor that contributed to these high values of manganese concentrations in the study areas.

Manganese ranges from 0.977-15.953 mg/L for boreholes water. All the values are higher than the standard values set by WHO (0.4 mg/L), USEPA (0.005 mg/L) and NSDWQ (0.2 mg/L). Manganese concentration ranges from 0.796-15.902 mg/L with a mean value of 8.011 mg/L for surface wells water. The values are all higher than the recommended standard by WHO (0.4 mg/L), USEPA (0.05 mg/L) and NSDWQ (0.2 mg/L).

The concentration of Nickel also ranges from 0.044-4.747 mg/L for boreholes water. The values exceeded the maximum permissible limit of concentration for drinking water by NSDWQ (0.02 mg/L), WHO (0.07 mg/L) and USEPA (0.1 mg/L) except OBA 4 which is lower than the reference level set by WHO (0.07 mg/L. The concentration of Nickel ranged from 0.011 – 7.835 mg/L for surface water, their concentration is low in OBA 2, OBA 8, OJC 1, than the WHO (0.07 mg/L).

Nickel high concentrations could be as a result of regular disposal of used batteries containing nickel by inhabitants in the study areas could result into such rise in contamination. The high value of this metal in the study areas could also be due to the presence of rocks and other minerals associated with nickel composition. From the geology of the study areas as indicated on the map, some of the sample areas are surrounded by hills characterised by several rocks and other mineral deposits. Nickel concentration is high at OBT 5, and low at OBA 2. The values in OBA 2, OBA 8, OJC 1, are lower than the standard by WHO (0.07 mg/L). All the other sample points have their values above the WHO (0.07 mg/L), NSDWQ (0.02 mg/L) and USEPA (0.1 mg/L).

Fortunately, the concentration of the heavy metal Lead (Pb), which is known to be very poisonous, has all its values falling below safe international standard limit. This suggests that the inhabitant of these study areas may be safe from the negative effect associated with excessive accumulation of Lead in an area. Subsequently one can say this affirm the safety on the use of surface well water in the study areas.

In this research work Table 1 shows the activities of alpha and beta for borehole water. The activities of alpha range from 0.004-0.081 Bq/L with a mean of 0.041 Bq/L and that of beta from 0.021-0.0174 Bq/L with a mean of 0.071 Bq/L. The mean values are both below the reference values of 0.5 Bq/L and 1.0 Bq/L by WHO (2011) for alpha and beta respectively. In a similar way, Table 5 shows that the gross alpha radioactivity for surface wells range from 0.002-0.054 Bq/L with a mean of 0.022 Bq/L and that of beta from 0.007-0.094 Bq/L and a mean of 0.058 Bq/L. Their values are below the reference acceptable values of 0.5 Bq/L for alpha and 1.0 Bq/L for beta by the WHO. The low values of alpha in this work could be stressed to low presence of limestone (sedimentary rocks), the use of fertilizers by farmers appears to have no adverse effects in this regards. The low values of gross beta activity in this investigation has no trace effect by the geological formation of the area through phosphorus fertilizers as reported to increase the gross beta in other works and also the presence of the sedimentary rocks in the areas. This confirmed no or low mining activities and low uranium, thorium and their progenies in these locations. This show that water source in this study areas are free from the radionuclides contamination.

6. Conclusion

This investigation reveals the heavy metals present water in the study area, their respective concentration, radioactivity concentration for gross Alpha, Beta and Annual Committed Equivalent Dose (CED) x-rayed by this work. It can be inferred that, the Borehole water and the hand dug well water in the study areas appeared safe for the dwellers and may have no significant health hazard on the people, however, there is need for constant water quality monitoring of the various sources of water as the results showed slightly high levels of some heavy metal concentration, signaling that a fraction of the populace may be at risk given the toxicity of these metals.

7. Limitation

Clearly this study is not without limitations, from the point of sample collection, apparatus used, preparation of the apparatus and the sample, collection of data, analysis of the data and the methods used, interpretations and representation. None has obvious source of error pointing avoidable limitations, however, exclusive efficiency of 100% isn't guarantee as known could constitute limitation.

8. Conflicts of Interest

The authors declare no conflict of interest.

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