Determination of Natural Radioactivity in Groundwater in Tanke-Ilorin, Nigeria

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Abstract

A study of the radioactivity in groundwater from Tanke-Ilorin, Nigeria, has been carried out. Ten water samples were analyzed by γ-ray spectroscopy to determine the ^{226}Ra and ^{228}Ra concentrations. The activity concentration values range from 0.81 ± 0.08 to 7.4 ± 2.2 Bq/l for ^{226}Ra and from 1.8 ± 0.3 to 5.6 ± 2.6 Bq/l for ^{228}Ra. The derived Annual Effective Dose received by the population as a result of the ingestion of ^{226}Ra was estimated to range from 0.08 ± 0.01 to 0.12 ± 0.07 mSv/y with an average of 0.39 ± 0.11 mSv/y and ^{228}Ra range from 0.50 ± 0.32 to 1.42 ± 0.70 mSv/y with an average of 0.91 ± 0.31 mSv/y. Consequently, the Annual Effective Dose received, as a result of the combined ingestion of ^{226}Ra and ^{228}Ra, was found to range from 0.81 to 1.74 mSv/y with an average of 1.30 mSv/y. The mean contribution of both ^{226}Ra and ^{228}Ra activities to the committed effective dose from a year's consumption of drinking water in the study area is, therefore, higher than the tolerable level of 1 mSv/y to the general public for prolonged exposure as recommended by ICRP, and much more than the new WHO recommended level of 0.1 mSv/y for drinking water.

Introduction

Water is vital and, concurrently, one of the most important natural resources. About 70% of the Earth's surface is covered with water, which is estimated at a volume of approximately 1.4 billion km^3 (Ashton *et al*., 2012). However, most of it is salty, and only around 2.5% of the global water resources (about 35 million km^3) consists of freshwater (merely a fraction of the freshwater can be used for drinking water purposes). Over 30% (about 10.5 million km^3) is stored as groundwater beneath the earth's surface – the most important available fresh water resource. Lakes and rivers account for far less than a half percent of the fresh water reserves. Groundwater is the most widespread and highly used resource; however, its quality can be endangered.

The increasing interest in radioactivity and its applications has brought about the need for an assessment of human exposure to radiation. It is, therefore, necessary to examine naturally occurring radioactivity in the environment, especially the occurrence of natural radioactivity in groundwater (Smith *et al*., 1961). Concentrations of dissolved radon gas provide one means of detecting the presence of natural radioactivity in groundwater, and, if the radioactivity level of groundwater is beyond tolerable limits, it could result into several health hazards among the populace (Arogunjo *et al*., 2004).

The increase in the level of radioactivity is as a result of possible migration of radionuclides to groundwater from anthropogenic activities: seepage of pollutants into the groundwater bodies, excessive fertilization of agricultural land, abandoned industrial sites, thermonuclear testing or nuclear power plants. Post-mining areas, waste dump areas or military areas could also contribute to the radioactivity increase. It is possible for groundwater to

naturally regenerate itself but, sometimes, takes many years as it proceeds very slowly.

Importance of water quality assessment and monitoring in the context of radioactivity cannot be overemphasized. Akinloye (1998) studied the radioactivity in a number of media, which include meat, fish, soil and water, as part of a pre-operational study of the nuclear facilities located in the Obafemi Awolowo University, Ile-Ife campus, Nigeria. The results of the studies showed that no man-made radionuclides were measured. The radionuclide concentration levels in surface soils in Ijero-Ekiti community (Ajayi *et al*., 1995) and soil/water samples obtained around production facilities of cement companies in Ewekoro and Port Harcourt cities in Nigeria (Avwiri, 2005; Jubril *et al*., 1999) were measured. Results from these studies did not reveal any significant level of radionuclides in the environment. More so, Nwankwo (2008, 2010) determined the levels of radioactivity in groundwater within the University of Ilorin permanent site but non-significant results were recorded. However, in a recent study of natural radioactivity of groundwater in Sango-Ilorin, the ingestion dose for some of the groundwater samples were above the new WHO recommended level of 0.1 mSv/y for drinking water but falls within the tolerable level of 1 mSv/yr to the general public for prolonged exposure as recommended by ICRP (Nwankwo, 2012).

Vengosh *et al.* (2009) reported that high levels of naturally occurring and carcinogenic radium isotopes have been measured in low-saline and oxic groundwater from the Rum Group of the Disi sandstone aquifer in Jordan. They revealed that the combined ^{228}Ra and ^{226}Ra activities in their study area are up to 2000% higher than international drinking water standards. The presence of high level of naturally occurring radionuclides in drinking water and food in a Taiwanese community was also reported and cited by Fasasi *et al*. (1999). Therefore, the call for this study in Tanke district, an area previously used as waste dumpsite and farmland is imperative. This would ascertain whether the level of radioactivity in the groundwater system could pose any significant health hazard to the populace.

Material and methods

Study area

The area of the study lies entirely within the basement rocks in the western part of central Nigeria and bounded by longitudes 4°36'–4° 39' E and latitudes 8°27'–8° 30' N (Fig. 1). This area falls within the northwestern part of Ilorin, a semi-arid region of Nigeria. The vegetation is mainly of the Guinea savanna type. Shrubs and underground rugged troughs and crests due to erosion characterized the topography of the area. The rocks are mainly bounded gneiss and auger gneiss with granodiorites and granites intrusions. The Nigeria basement complex (Fig. 2) consists of at least four main groups of rocks: the migmatite gneiss complex, the metasediment (composed of schist, calc-gneiss, quartizite and meta-conglomerate), the porphyritic older granite and the miscellaneous rock types, which are mostly post orogenic rocks like aplite, pegmatites, and dolerites dykes. These rocks are younger and are found to cut through pre-existing rocks (Rahaman, 1973).

Samples collection and treatment

Water samples were collected from 10 hand dug wells scattered within Tanke community in Ilorin, Nigeria. The locations are shown in Fig. 1 and Table 1. About 6

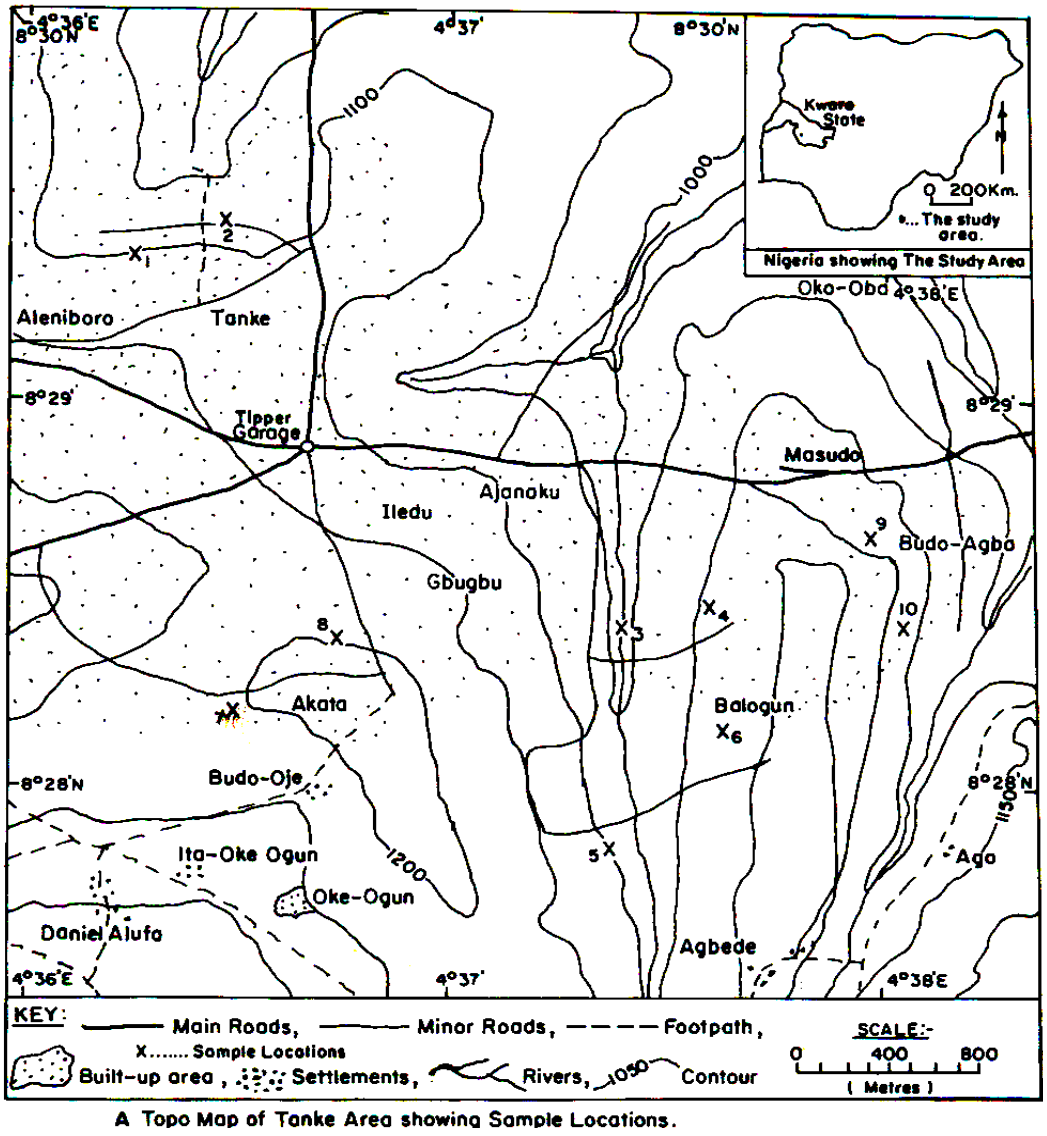
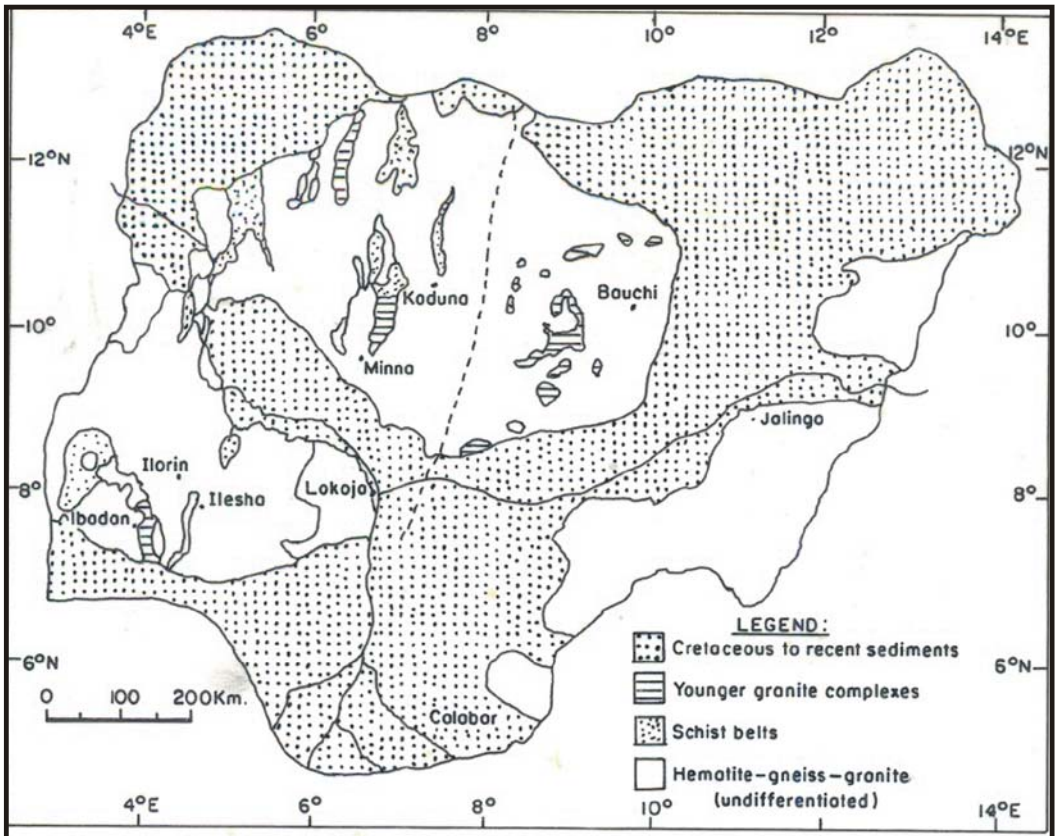


Fig. 1. Topographical map of the study area showing sample locations

litres of water were collected in polyethylene containers. The water samples were acidified with 11 *M* of H_3O^+, Cl^- at the rate of 10 ml per litre of sample as soon as possible after sampling to avoid absorption of radionuclides on the walls of the containers (IAEA, 1986; Lydie & Nemba, 2008). Marinelli beaker of 1 litre volume capacity, previously washed, rinsed with a dilute sulphuric acid and dried to avoid contamination, were filled with known volume of the various water samples and firmly sealed. The sealed water samples



A geological map of Nigeria

Fig. 2. Geological map of Nigeria showing Ilorin City

TABLE 1
Locations of the groundwater samples in the study area

S/N	*Location of samples*	*Code*
1	Tanke Ilewe I	X_1
2	Tanke Ilewe II	X_2
3	Tanke Ajanaku I	X_3
4	Tanke Ajanaku II	X_4
5	Tanke Bubu I	X_5
6	Tanke Bubu II	X_6
7	Akata Tanke I	X_7
8	Akata Tanke II	X_8
9	Tanke Oke-Odo I	X_9
10	Tanke Oke-Odo II	X_{10}

were subsequently stored in a refrigerator and kept for 30 days to ensure that no loss of radon occurs, thereby, ensuring a state of secular equilibrium to be reached between radium isotopes and their respective daughters before γ-ray spectrometry analysis was carried out. This method has been commonly used by workers (Lydie & Nemba, 2008; Jubril *et al*., 1999; Ajayi *et al*., 1995). The spectrometer consists of a Canberra 7.6 cm by 7.6 cm NaI (TI) detector coupled with a Canberra series 10 plus Multichannel Analyzer (MCA) through a pre-amplifier base. Environmental shielding of

the water was achieved using a Canberra 10 cm thick lead castle (Farai & Sanni, 1992).

Counting was done for 10 hr because of the low natural activities of radionuclides in water. The spectrum was measured and the area under the photopeaks was computed using the algorithm of the MCA. The prominent photopeaks observed in the spectra of the samples were identified as those of the radionuclides in the natural decay series of ^{238}U and ^{232}Th, and the non-series ^{40}K. Subsequently, the transition lines of 1764.5 keV of ^{214}Bi and 2614.7 keV of ^{208}Tl were used to determine the concentrations of non-series nuclide ^{226}Ra (decay series of naturally occurring radionuclide headed by ^{238}U) and ^{228}Ra (decay series of naturally occurring radionuclide headed by ^{232}Th), respectively. Spectra analysis software, SAMPO 90, was used to match various gamma energy peaks to library of possible radionuclides (Ahmed, 2004; Nasirian *et al.*, 2008; Lydie & Nemba, 2009; Ajayi & Adesida, 2009; Nwankwo, 2012). The non-series ^{40}K were not considered in this study (explained later in the paper).

Radioactivity computation

Each radionuclide concentration C, in each water sample was evaluated using the relation (Lydie & Nemba, 2009):

$$C = \frac{N(E_y)}{\varepsilon(E_y).I_y.Vt_c} \qquad (1)$$

where $N(E_y)$ is the net peak area of the radionuclide of interest, $\varepsilon(E_y)$ is the efficiency of the detector for the energy E_y, I_y is the intensity per decay for the energy E_y, V is the volume of the water sample and t_c is the total counting time in seconds (36000 s).

When analyzing the annual effective dose to the human population from natural sources, the dose received by ingestion of long-lived natural radionuclides must be considered. Effective doses resulting from the intake of ^{226}Ra and ^{228}Ra may be determined directly from external measurements of their concentrations in the body or estimated from concentrations intake materials such as air, food and water (Ahmed, 2004). Intakes of the natural radionuclides ^{226}Ra and ^{228}Ra through groundwater in Tanke-Ilorin were calculated. The annual effective dose was calculated with the intake of individual radionuclide and ingestion dose coefficients (Sv Bq^{-1}) reported by the International Commission on Radiological Protection (ICRP, 1994). The equation for calculating the annual effective dose (AED) per person is given by (Ahmed, 2004; Ajayi & Adesida, 2009; Nwankwo, 2012):

$$AED = \sum_i I_i.365.D_i \qquad (2)$$

where I_i is the daily intakes of radionuclide I (Bq d^{-1}) and the ingestion dose coefficient D_i for ^{226}Ra and ^{228}Ra is 2.8×10^{-7} and 6.9×10^{-7} SvBq^{-1}, respectively (ICRP, 1994).

Results and discussion

The radionuclides identified in the water samples and quantified from the γ ray spectra are ^{226}Ra and ^{228}Ra, which are the decay daughter products of naturally occurring radioactive elements ^{238}U and ^{232}Th. The activity of naturally occurring radionuclides for the 10 different locations is shown in Table 2. The activity concentration values range from 0.81 ± 0.08 to 7.4 ± 2.2 Bq l^{-1} with an average of (3.7 ± 1.1) Bq l^{-1} for ^{226}Ra and from 1.8 ± 0.3 to 5.6 ± 2.6 Bq l^{-1} with an average of (3.6 ± 1.2) Bq l^{-1} for ^{228}Ra.

TABLE 2
The mean specific activity concentration of the water samples

Sample	^{226}Ra *(Bql^{-1})*	^{228}Ra *(Bql^{-1})*
X_1	2.4 ± 0.4	5.1 ± 1.2
X_2	3.8 ± 1.3	3.2 ± 1.3
X_3	0.8 ± 0.1	5.6 ± 2.6
X_4	7.4 ± 2.2	1.8 ± 0.3
X_5	5.3 ± 1.1	2.3 ± 0.1
X_6	4.1 ± 1.2	3.2 ± 0.7
X_7	6.2 ± 2.4	4.4 ± 1.8
X_8	5.0 ± 1.5	2.1 ± 0.8
X_9	1.1 ± 0.7	5.1 ± 2.6
X_{10}	1.1 ± 0.2	2.8 ± 1.7
Mean	3.7 ± 2.2	3.6 ± 1.3

High levels of ^{40}K were also detected in the groundwater samples, which are function of the geological formation of the area (Watson, 1986). ^{40}K is the principal naturally occurring source of internal radiation despite its low isotopic abundance (IAEA, 1986). However, because it is an essential biologic element, which is under close metabolic control, variations in dietary composition have little effect on the body content or on the radiation dose received (NCRP, 1967). The mean concentration of ^{226}Ra is relatively low and agreed with a range of groundwater values obtained by some investigators, namely Ziqiang *et al*. (1988): up to 4 Bq l^{-1}; Tchoskossa (1998): 7–16 Bq l^{-1}. The ^{228}Ra recorded by Tchoskossa (1998) was between 2.3–4.0 Bq l^{-1}. Table 3 shows the average values for ^{226}Ra and ^{228}Ra concentration in groundwater samples of investigation in some other countries.

Assuming the volume of drinking water for adult to be 1 litre/day (WHO, 1993; Ahmed, 2004; Lydie & Nemba, 2009) in the study area, the daily intake per person of ^{226}Ra and ^{228}Ra through groundwater in all locations and the AED are presented in Table 4. The derived AED received by the population as a result of the ingestion of ^{226}Ra is estimated to range from 0.08 ± 0.01 to 0.12 ± 0.07 mSv y^{-1} with an average of 0.39 ± 0.11 mSv y^{-1} and ^{228}Ra range from 0.50 ± 0.32 to 1.42 ± 0.70 mSv y^{-1} with an average of 0.91 ± 0.31 mSv y^{-1}. The AED received

TABLE 3
Average values for ^{226}Ra and ^{228}Ra concentration in groundwater samples of investigation in some other countries (modified after Ahmed (2004) and Lydie &Nemba (2009)).

Country	^{226}Ra	^{228}Ra	*Reference*
China	0.34	–	*
China	Up to 4	0.3	Ziqiang *et al*. (1988)
Poland	1.1	–	**
Egypt (Qena)	2.1	1.1	Ahmed (2004)
(Safaga-Qusier)	3.1	1.4	Ahmed (2004)
Nigeria (Ife)	7.0	3.3	Tchokossa (1998)
Nigeria (Tanke- Ilorin)	3.7	3.6	Present work

* Weihai Z., Takao I. and Xiaotang Y. (2001). Occurrence of Rn-222, Ra-226, Ra-228 and U in Groundwater in Fujia Province, China, *J. Envir. Radio*., 53, 111–120.

** Jankowski J., Chruscielewski W., Kaminski Z. and Zak A. (2000). Natural Radioactivity of Underground Water Supplies in the Region Lodz in Poland", IRPA 10. Scientic topics-1 Natural Radiation Exposure.

TABLE 4
Daily intake of ^{226}Ra and ^{228}Ra and the estimated annual effective doses from the water samples

Sample	*Intake per person (Bq d^{-1})*		*Annual effective dose (mSv y^{-1})*		*Annual effective dose (combined) (mSv y^{-1})*
	^{226}Ra	*^{228}Ra*	*^{226}Ra*	*^{228}Ra*	
X_1	2.4 ± 0.4	5.1 ± 1.2	0.24 ± 0.04	1.30 ± 0.30	1.54
X_2	3.8 ± 1.3	3.2 ± 1.3	0.40 ± 0.13	0.80 ± 0.32	1.2
X_3	0.8 ± 0.1	5.6 ± 2.6	0.08 ± 0.01	1.42 ± 0.70	1.5
X_4	7.4 ± 2.2	1.8 ± 0.3	0.80 ± 0.23	0.50 ± 0.07	1.3
X_5	5.3 ± 1.1	2.3 ± 0.1	0.60 ± 0.11	0.60 ± 0.03	1.2
X_6	4.1 ± 1.2	3.2 ± 0.7	0.42 ± 0.12	0.81 ± 0.12	1.23
X_7	6.2 ± 2.4	4.4 ± 1.8	0.64 ± 0.24	1.10 ± 0.50	1.74
X_8	5.0 ± 1.5	2.1 ± 0.8	0.52 ± 0.14	0.53 ± 0.07	1.05
X_9	1.1 ± 0.7	5.1 ± 2.6	0.12 ± 0.07	1.30 ± 0.70	1.42
X_{10}	1.1 ± 0.2	2.8 ± 1.7	0.11 ± 0.02	0.70 ± 0.32	0.81
Mean	3.7 ± 2.2	3.6 ± 1.3	0.39 ± 0.11	0.91 ± 0.31	1.3

as a result of the combined ingestion of ^{226}Ra and ^{228}Ra is, consequently, found to range from 0.81 to 1.74 mSv y^{-1} with an average of 1.30 mSv y^{-1}.

In the area under investigation, the AED received as a result of the combined ingestion of ^{226}Ra and ^{228}Ra from each of the sampled groundwater are above 1 mSv y^{-1}, except for sample X_{10}. According to ICRP recommendations (ICRP, 1991), the public should not be exposed to more than an average of 1 mSv y^{-1}, while in WHO's recent publication, 0.1 mSv y^{-1} (WHO, 2000) is recommended for drinking water. The average AED received by the populations of Tanke-Ilorin area, as a result of the combined ingestion of ^{226}Ra and ^{228}Ra in groundwater, is, therefore, above these tolerable limits.

Conclusion

The study of the radioactivity of groundwater in Tanke district, Ilorin Kwara State has been carried out. The mean value has been taken as the estimated value for the area. The derived AED received by the population as a result of the ingestion of ^{226}Ra is estimated to range from 0.08 ± 0.01 to 0.12 ± 0.07 mSv y^{-1} with an average of 0.39 ± 0.11 mSv y^{-1} and ^{228}Ra range from 0.50 ± 0.32 to 1.42 ± 0.70 mSv y^{-1} with an average of 0.91 ± 0.31 mSv y^{-1}; while the AED received by the combined ingestion of ^{226}Ra and ^{228}Ra is, consequently, found to range from 0.81 to 1.74 mSv y^{-1} with an average of 1.30 mSv y^{-1}. The mean contribution of ^{226}Ra and ^{228}Ra activities to the committed effective dose from a year's consumption of drinking water in the study area is, therefore, 30% higher than the tolerable level of 1 mSv y^{-1}to the general public for prolonged exposure as recommended by ICRP, and more than 1000% above the new WHO recommended level of 0.1 mSv y^{-1} for drinking water.

The study shows that it may not be radiologically safe to consume the groundwater from the sampled area since they are above the recommended limit. In view of the fact that measurements of natural

radioactivity in drinking water is performed mostly for assessment of the doses and risk resulting from consuming water, consumption of the groundwater in the area may pose health side-effects to the public. It is, therefore, recommended that appropriate measures should be made to protect the populace from adverse health implication. It is also recommended that such study be routinely carried out in boreholes, wells and even tap water at least bi-annually as a check.

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