International Journal of the Physical Sciences Vol. 5(7), pp. 1009-1014, July 2010
Available online at http://www.academicjournals.org/IJPS
ISSN 1992 - 1950 ©2010 Academic Journals

Full Length Research Paper

Chemical and radiological risk assessment of uranium in borehole and well waters in the Odeda Area, Ogun State, Nigeria

C. M. Amakom* and N. N. Jibiri

Radiation and Health Physics Research Laboratory, Department of Physics,
University of Ibadan, Ibadan, Nigeria.

Accepted 27 May, 2010

The activity concentrations of uranium in well and borehole water samples in the Odeda area were measured using gamma ray spectroscopic system. The results obtained were used to calculate human radiological risk over lifetime consumption by the population in the area. The activity concentrations of uranium in all the water supplies for drinking and domestic purposes were found to range from 0.51 ± 0.02 to 6.77 ± 0.23 Bq. l^{-1}. The mass concentration was found to range from 20.17 ± 0.79 to 267.80 ± 9.09 µg.l^{-1}. The radiological risks for cancer mortality risk were found to be low, typically ranging from 1.91 × 10^{-5} to 2.54 × 10^{-4}, while that of the morbidity risk ranged from 2.93 × 10^{-5} to 3.89 × 10^{-4}. However, the chemical toxicity was found to vary from 0.56 – 7.47 µg.kg^{-1}.day^{-1} which is higher than the recommended acceptable safe level by various international organizations. Results of measurements could be of vital importance in radio-epidemiological assessment, diagnosis and prognosis of uranium induced diseases in the local population of the area under investigation.

Key words: Uranium, radiological risk, toxicity risk, cancer mortality and morbidity risk.

INTRODUCTION

Uranium salt is the most soluble of the long-lived radionuclides and forms ions with oxidation states of +4 (UO_2 and U^{4+}) and +6 (UO_3 and UO_2^{2+}) (Salonen, 1994; Banks et al., 1995). Uranium will bond with oxygen to form the uranyl ion, or uranium dioxide, which is soluble in ground water under aerobic conditions. In Nigeria, clean water for drinking and domestic uses in rural areas has become an uphill task to achieve and probably could be seen a luxury. This could be attributed to high poverty rate of about 54.6% in the country (NLSS, 2005; NBS, 2008) and also, due to constant decline of portable water in the rural areas since the 1990s (WHO, 2003). In the Odeda area where this work was carried out, the major source of drinking and domestic water supply to the local population is ground water and more often during the rainy seasons people relied on rain water collections from the roofs of their houses. Previous studies in the study areas showed that the area is associated with high outdoor gamma radiation exposure levels, with typical range and mean of the total absorbed dose rates due to primordial radionuclides (^{238}U, ^{232}Th and ^{40}K) being 0.035 to 0.328 and 0.214 ± 0.049 µGy.h^{-1}, respectively (Jibiri et al., 1999; Farai and Jibiri, 2000; Jibiri, 2001; Farai and Vincent, 2006). It has been established that high concentrations of uranium greater than 15 µg.l^{-1} in domestic water may present harmful biological effects in humans (WHO, 2008). The toxic effects of uranium compounds have been extensively studied in kidney (Guglielmotti et al., 1989; Leggett, 1989; Kutttio et al., 2002; Miriam et al., 2009) and bone of laboratory animals (Larivière et al., 2007). The chemical toxicity effects on

*Corresponding author. E-mail: camakom@gmail.com, chijiokemichael@yahoo.com. Tel: +234-08037779552.

the human kidney by chronic ingestion of uranium through drinking water in the range of 0.004 to 9 µg.l^{-1} per body weight per day may produce interference with kidney functions (Zamora, 1998). In a more recent study on humans by Kurttio et al. (2005), nephrotoxic effects of uranium in drinking water were found even for low concentrations – without a clear threshold. Most results from uranium studies in drinking water suggest that the safe concentration of uranium in drinking water may be within the range of proposed guideline values of 2 - 30 µg.l^{-1} (Kurttio et al., 2002; WHO, 2008). Because uranium is a predominantly alpha-emitting radionuclide, there is a concern about the potential DNA damage if the emitted alpha particles reach the cell nuclei, of the body for instance through water ingestion. Attempts by cells to repair this damage, if it occurs, may result in repair errors, producing gene mutations or chromosomal aberrations. These effects, when sufficiently severe, may be manifested as cancer and possibly as developmental malformations in children and developing foetus. There are little or no data on the radiological and chemical health effects of uranium in drinking water and domestic supplies in Odeda area of Ogun state, Nigeria despite the characterization of the area by agricultural and stone quarrying activities while hand dug wells and boreholes are the major sources of water. Hence, ground water can become contaminated by domestic sewage, feedlots and surface runoff, as well as other pollution sources such as quarrying activities. Where the sub-surface geology permits rapid downward movement of water sources from the surface or where ground and well water sources are tapped near the surface, aquifers may be largely affected (Banks et al., 1995). This study was considered relevant in order to provide data on the concentrations of uranium in the borehole and well waters and on the associated radiological and chemical risks it may pose to the population in Odeda Area which is known for granitic quarry activities.

MATERIALS AND METHODS

Sampling

In order to ensure that sampling efforts relatively covers the study area, a pre-field survey was made to identify sampling locations for borehole water and hand dug wells. These identified water locations are those that are largely used by the local population. A total of three (3) hand-dug well and four (4) borehole water supplies were selected. The water samples were collected with 1 L-size plastic containers which has been previously washed and rinsed with dilute acid (0.1 M HCl). The collected samples were acidified with 1 M concentrated HCl to obtain a pH < 2 in order to avoid adsorption of the radionuclide on the walls of the container (AS/NZS, 1998). The samples were subsequently taken to the laboratory for preparation prior to gamma spectroscopy. Figure 1 shows the map of the state and locations of the sampling points in the study area where the samples were collected.

Measurement

The method employed for the measurements of the radioactivity in the samples was the gamma-ray spectroscopy and the standard procedures of this method as described in literatures were followed (Itsumusa and Kousuke, 1984; Awudugba and Techokossa, 2008) and have been employed in our previous studies (Jibiri et al., 1999, Jibiri et al., 2007, 2009). The detector used for the radioactivity measurements is a lead-shielded 76 x 76 mm NaI(Tl) detector crystal (Model No. 802 series, Canberra Inc.) coupled to a Canberra Series 10 plus Multichannel Analyzer (MCA) (Model No.1104) through a preamplifier. It has a resolution Full Width at Half Maximum (FWHM) of about 8 % at energy of 0.662 MeV (^{137}Cs) which is considered adequate to distinguish the gamma ray energies of interest in this study. The choice of radionuclides to be detected was predicted based on the fact that the NaI(Tl) detector used in this study had a modest energy resolution. Hence the photons emitted by them would only be sufficiently discriminated if their emission probability and their energy are high enough and the surrounding background continuum was low enough. Therefore, the activity concentration of ^{214}Bi (determined from its 1.760 MeV γ -ray peak) was chosen to provide an estimate of ^{226}Ra (^{238}U) in the samples. The samples were placed symmetrically on top of the detector and measured for a period of 10 hours. The net area under the corresponding peaks in the energy spectrum was computed by subtracting counts due to Compton scattering of higher peaks and other background sources from the total area of the peaks. From the net area, the activity concentrations in the samples were obtained using (Jibiri et al., 1999; Awudugba and Techokossa, 2008):

$$C\left(Bq\ .l^{-1}\right) = kC_n \quad (1)$$

Where $k = \frac{1}{\varepsilon P_\gamma V_s}$, C is the activity concentration of the radionuclide in the sample given in Bq l^{-1}, C_n is the count rate under the corresponding peak, ε is the detector efficiency at the specific γ-ray energy, P_γ is the absolute transition probability of the specific γ-ray energy, and V_s is the volume of the sample (l). The detailed calibration procedures of the detector can be found in our publications (Jibiri et al., 1999, Jibiri et al., 2007, 2009). Using equation 1, the activity concentrations of uranium in each of the water samples were determined.

RESULTS AND DISCUSSION

Activity and mass concentration determination

The result of the activity and mass concentrations of Uranium in each of the samples are presented in Table 1. As could be seen in Table 1 the activity concentration of uranium ranged from 2.58 ± 0.09 to 3.24 ± 0.33 Bq.l^{-1} for the hand dug well waters while for the borehole waters it ranged from 0.51 ± 0.02 to 6.77 ± 0.23 Bq.l^{-1}. The error in these reported values are combined uncertainties in the

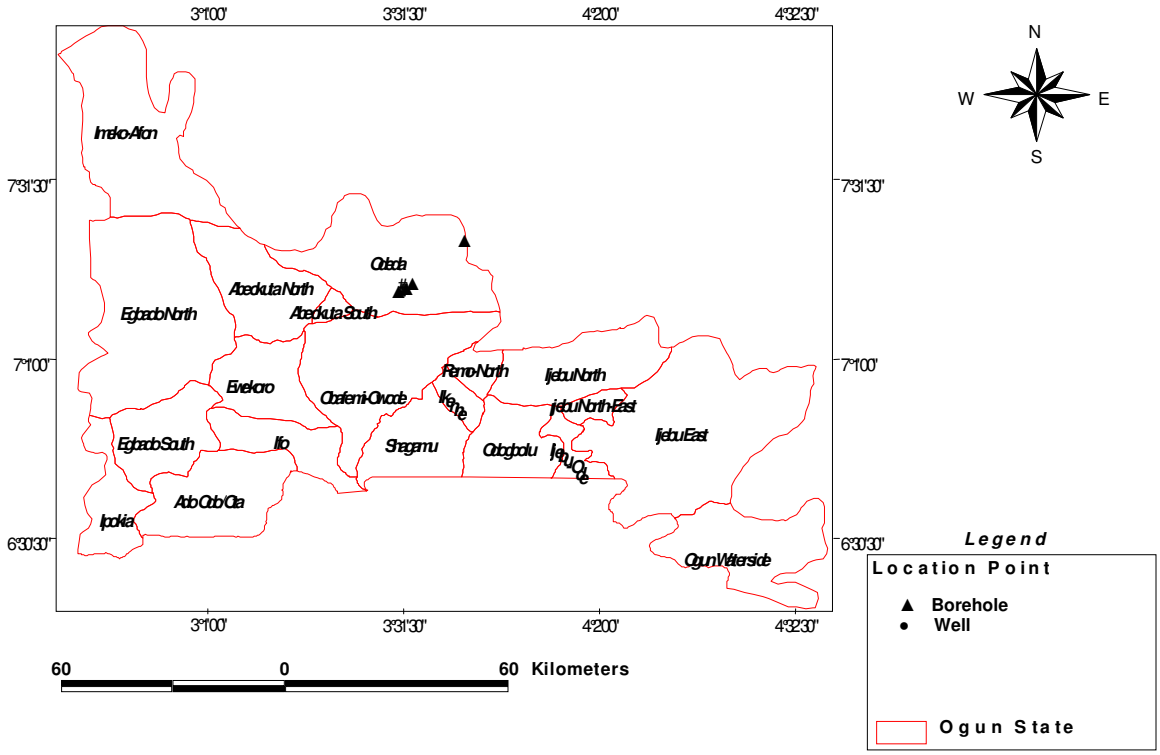

Figure 1. Map of Ogun State showing GPS points of sample collections.

Table 1. The activity concentrations and mass concentration of Uranium in the dug well (WL) and borehole (BH) water samples in the study area.

Sample	^{238}U (Bq.L^{-1})	^{238}U (pCi.L^{-1})	^{238}U (µg.L^{-1})
WL 1	2.58 ± 0.09	69.66 ± 2.43	103.97 ± 3.62
WL 2	3.65 ± 0.04	98.55 ± 1.08	147.08 ± 1.61
WL 3	3.24 ± 0.33	87.48 ± 8.91	130.56 ± 13.29
BH 1	5.54 ± 0.19	149.58 ± 5.13	223.25 ± 7.65
BH 2	6.77 ± 0.23	182.79 ± 6.21	272.82 ± 9.26
BH 3	0.51 ± 0.02	13.77 ± 0.54	20.55 ± 0.80
BH 4	5.01 ± 1.27	135.27 ± 34.29	201.89 ± 51.17

Hand dug well water sample (WL), Borehole water sample (BH).

counting error, efficiency determination etc. The data for the activity concentrations of the uranium were converted to the uranium mass concentration (µg.l^{-1}) using the following conversion factors:

$$1Bq\,.l^{-1} = 27.0\,pCi\,.l^{-1}; 1\mu gl^{-1} = \frac{1\,pCi\,.l^{-1}}{0.67} \quad (2)$$

The mass concentration values are also presented in Table 1. As could be seen, it ranged from 20.55 ± 0.80 to 272.82 ± 9.26 µg.l^{-1} for the samples. Various health and environmental protection agencies have recommended a safe limit of uranium in drinking water for human beings. The World Health Organization (WHO, 2003) has recommended 15 µg.l^{-1} of uranium in water as a safe limit for drinking purposes. The United States Environment Protection Agency (USEPA, 1994, 2003) has regulated uranium in public water supplies and has set the value of 30 µg.l^{-1}as the safe limit. Health Canada (1999) sets the standard as 20 µg.l^{-1}. These levels were set to represent

the concentration that would not result in any significant risk to health over a lifetime's drinking of water. Comparing uranium concentrations in the various samples obtained in this study with the recommended values of 15 $\mu g.l^{-1}$ (WHO, 2008), 20 $\mu g.l^{-1}$ (Health Canada and Australia, 2000), and 30 $\mu g.l^{-1}$ (USEPA, 2003), it could be seen that only sample BH 3 had a value about the USEPA safe limit while others exceeded the limits. These high values may be attributed to the fact that the area is known to have high outdoor gamma radiations to which major contributor to dose rates from uranium and thorium decay series (Farai and Jibiri, 2000; Jibiri, 2001).

Radiological risk assessment

The lifetime cancer risks, R, associated with intake of a given radionuclide were estimated from the product of the applicable risk coefficient *r* and the per capita activity intake I expressed in equation (3).

$$R = r \times I \quad (3)$$

According to WHO (2008), the average life expectancy at birth in Nigeria is 45.5 y and, an annual consumption of water for an individual is about 730 l. This brings the lifetime intake of water to 33,215 l. The cancer risk coefficients of uranium of 1.13×10^{-9} and 1.73×10^{-9} Bq^{-1} for mortality and morbidity respectively were obtained from the literature (EPA, 1999; UNSCEAR, 2000). Using equation 3 and these coefficients the cancer mortality and morbidity risks of uranium over lifetime consumption of water were calculated. The results are presented in Table 2. From the table the cancer mortality risk ranged from 1.91×10^{-5} to 2.54×10^{-4} while for morbidity risk, it ranged between 2.93×10^{-5} and 3.18×10^{-4}. The cancer risk at 10^{-4} is low compared to the acceptable level of 10^{-3} for the radiological risk (Ye-shin et al., 2004).

Chemical toxicity risk

The chemical toxicity risk was evaluated using the lifetime average daily dose of uranium through drinking water intake, and compared it with the reference dose (RFD) of 0.6 $\mu g.kg^{-1}.day^{-1}$ (Ye-shin et al., 2004) used as a standard criteria for uranium in several foreign organizations and thereby produce a hazard quotient (Equation 4).

$$Hazard\ quotient = \frac{LADD}{RFD} \quad (4)$$

Table 2. The estimated lifetime cancer mortality and morbidity risk of uranium in the water samples.

Sample	Cancer mortality risk	Cancer morbidity risk
WL 1	9.68×10^{-5}	1.48×10^{-4}
WL 2	1.36×10^{-4}	2.09×10^{-4}
WL 3	1.21×10^{-4}	1.86×10^{-4}
BH 1	2.07×10^{-4}	3.18×10^{-4}
BH 2	2.54×10^{-4}	3.89×10^{-4}
BH 3	1.91×10^{-5}	2.93×10^{-5}
BH 4	1.88×10^{-4}	2.87×10^{-4}

Hand dug well water sample (WL), Borehole water sample (BH).

$$Ingestion\ LADD\ of\ drinking\ water = \frac{EPC \times IR \times EF \times ED}{AT \times BW} \quad (5)$$

Where LADD, lifetime average daily dose ($\mu g.kg^{-1}.day^{-1}$); EPC is the exposure point concentration ($\mu g.l^{-1}$); IR is the water ingestion rate ($l.day^{-1}$); EF is the exposure frequency ($days.year^{1}$); ED is the total exposure duration (years); AT is the average time (days) and BW is the body weight (kg). Using therefore, IR = 2 $l.day^{-1}$; EF = 350 days, ED = 45.5 y, AT = 16,607.5 (obtained from 45.5 × 365) and BW = 70 kg (for a standard man) the chemical toxicity risk of uranium over a lifetime consumption was estimated. The result is presented in Table 3. The exposure dose ranged from 0.56 - 7.47 $\mu g.kg^{-1}.day^{-1}$. The LADDs values were observed higher in the borehole samples than in the hand dug well water samples. This could be due to the depth of the water source, underground water and the geochemistry when compared to the hand dug well of depths not greater than 30 m. By comparing the lifetime average daily dose (LADD) obtained in this study and the reference dose (RFD) (0.6 $\mu g.kg^{-1}.day^{-1}$) that is an acceptable level, the chemical toxicity risk due to uranium in the water samples were all above the RFD.

This shows that there are health risks associated with uranium in the water samples which are mainly due to the chemical toxicity risk.

Conclusion

The activity concentration of uranium in hand dug well and borehole water samples in Odeda Area of Ogun State Nigeria have been carried out using gamma-ray spectroscopy. The activity concentrations were related to the mass concentrations of uranium in the samples. The mass concentration was found to vary from 20.55 to 272.82 $\mu g.l^{-1}$. The results of measurement indicate that the measured mass concentrations of uranium in the well

Table 3. The estimated lifetime average daily dose (LADD) of uranium in the water samples.

Sample	LADD ($\mu g.kg^{-1}.day^{-1}$)
WL 1	2.84
WL 2	4.02
WL 3	3.57
BH 1	6.11
BH 2	7.47
BH 3	0.56
BH 4	5.53

and borehole water supplies in the area were relatively high when compared with the recommended safe limits by some various international organizations. The radiological risks of uranium in the water samples were found to be low, typically in magnitude of 10^{-4}. It could therefore be inferred that the human risk due to uranium content in water supplies that will result from ingestion in the area may likely be to the chemical toxicity of uranium as a heavy metal rather than radiological risk. However, this study represents small area coverage, but it suffices to say that it represents a useful radiometric data that could be of vital importance in radio-epidemiological assessment, diagnosis and prognosis of uranium-induced diseases to the local population in of the area under investigation.

REFERENCES

Australia and New Zealand Standards (AS/NZS) (1998). Water quality – Sampling Guidance on the design of sampling programs, sampling techniques and the preservation and handling of samples (AS/NZS 5667-1).

Awudugba AO, Tchokossa P (2008). Assessment of Radionuclide Concentrations in Water Supply from Bore-holes in Ogbomoso land, Western Nigeria; Ind and Buil. Environ., 17(2): 183-186.

Banks D, Royset O, Strand T, Skarphagen H (1995). Radioelement (U, Th, Rn) concentrations in Norwegian bedrock groundwaters: Environ. Geol., 25: 165-180.

Environmental Protection Agency (EPA) (1999). Cancer risk coefficients for Environmental exposure to radionuclides. United State Environmental Protection Agency. Federal Guidance Report No -13 (EPA. 402 R-99-001).

Farai IP, Jibiri NN (2000). Baseline studies of terrestrial outdoor gamma dose rate levels in Nigeria: Radiation Protection Dosimetry, 88: 247 - 254.

Farai IP, Vincent UE (2006). Out-door radiation level measurement in Abeokuta, Nigeria, by thermoluminescent dosimetry; Nig. J. Phys., 18(1): 121-126.

Guglielmotti M, Ubios A, Larumbe J, Cabrini R (1989). Tetracycline in uranyl nitrate intoxication: its action on renal damage and U retention in bone. Health Phys., 57: 403-405.

Health Canada (1999). Summary of guidelines for Canadian drinking water quality: Prepared by federal provincial subcommittee on drinking water of the federal provincial-territorial committee on environmental and occupational health.

Itsumusa U, Kousuke K (1984). Feasibility Study on a continuous environmental gamma-ray monitoring using a NaI(Tl) scintillation spectrometer. Health Phys., 33: 559-568.

Jibiri NN (2001). Assessment of health risk levels associated with terrestrial gamma radiation dose rates in Nigeria; Environ. Inter., 27(1): 21-26.

Jibiri NN, Alausa SK, Farai IP (2009). Assessment of external and internal doses due to farming in high background radiation areas in old tin mining localities in Jos-Plateau, Nigeria. Radioprotection, 44: 139-151.

Jibiri NN, Farai IP, Alausa SK (2007) Estimation of annual effective dose due to Natural radioactive elements in ingestions of foodstuffs in tin mining area of Jos-Plateau, Nigeria: J. Environ. Radioactivity, 94: 31- 40.

Jibiri NN, Mabawonku AO, Oridate AA, Ujiagbedion CA (1999). Natural radionuclide concentration levels in soil and water around a Cement factory at Ewekoro, Ogun, Nigeria: Nig. J. Phy., 11: 12-16.

Kurttio P, Auvinen A, Salonen L (2002) Renal Effects of Uranium in Drinking Water Environmental Health Perspective. 110(4): 337-342.

Kurttio P, Komulainen H, Leino A, Salonen L, Auvinen A, Saha H (2005). Bone as a possible target of chemical toxicity of natural uranium in drinking water. Environmental Health Perspective. 113: 68-72.

Larivière D, Packer AP, Mario L, Li C, Chen J, Cornett RJ (2007). Age dependence of natural uranium and thorium concentrations in bone. Health Phys., 92: 119-126.

Leggett R (1989). The behavior of and chemical toxicity of U in the kidney: A Reassessment Health Phys., 57: 365-383

Miriam SK, Carolina MT, Maria CM, Juan CF (2009). Uranium in drinking water: effects on Mouse oocyte quality. Health Phy., 96: 568-574.

National Bureau of Statistics (NBS) (2008). Annual abstract of statistics, Abuja.

Nigerian Living Survey Standard (NLSS) (2005). Report of the Nigerian living standard survey, Abuja National Planning Commission 2.

United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR) (2000). Sources, effects and risks of ionizing radiation. Report to the General Assembly. ISBN 92-1-142238-8, New York.

United State Environmental Protection Agency (USEPA) (1994). Comparison of Health Risks Arising From the Radiological and Chemical Toxicity of Uranium**,** Appendix F of Radiation Site Cleanup Regulations: Technical Support Document For The Development Of Radionuclide Cleanup Levels For Soil, Review Draft*,* EPA 402-R-96-011 A.

United States Environmental Protection Agency (2003). Current Drinking Water Standards*,* Ground water and drinking water protection agency, pp. 1–12.

World Health Organization (WHO) (2003). Guidelines for drinking water quality Health criteria and other supporting information, Geneva, 2nd edn., 2: 367–370.

World Health Organization (WHO) (2008). Meeting the MDG drinking water and sanitation target: the urban and rural challenge of the decade. WHO Library Cataloguing-in-Publication Data.

Ye-shin K, Hoa-sung P, Jin-yong K, Sun-ku P, Byong-wook C, Ig-hwan S, Dong-Chun S (2004). Health risk assessment for uranium in Korean groundwater. J. Environ. Radioactivity, pp. 77-85.

Zamora ML, Tracy BL, Zielinski JM, Meyerhof DP, Moss MA (1998). Chronic Ingestion of Uranium in Drinking Water: A Study of Kidney Bioeffects in Humans, Toxicol. Sci., 43: 31 -45.