

Nigerian Journal of Physics (NJP)

ISSN: 1595-0611

Volume 32(1), March 2023



Environmental Impact Assessment of Radionuclide and Metal Contamination in Heavily Urbanized River, Ibadan, Nigeria

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ABSTRACT

Naturally occurring radionuclides and other contaminants are present in drinking water, these contaminants are threats to human health. The present study measured activity concentrations of (40K, 228Ra, and 232Th) and some heavy metals in Ona River, Ibadan, Southwestern Nigeria. Fifteen water samples were collected from different points at different locations along the course of the river. The activity concentrations and heavy metals were analyzed using NaI(Tl) gamma-ray spectrometer and Atomic Absorption Spectrometer (AAS). The average values for 40 K, 226 Ra and 232 Th were 21.89 Bq·L⁻¹, 5.53 Bq·L⁻¹, 3.87 Bq·L⁻¹ respectively. Lifetime cancer risk obtained in the present study as compared with the world recommendation showed no elevated risk of lifetime cancer due to the ingestion of the water. The mean annual effective dose for adults in the present study was not different from the WHO standard but the mean annual effective dose value for children in the present study is two times higher than the WHO standard values. The total average effective dose for children was also higher than the world recommended value contrary to adults, estimated total dissolved solids (TDS) vary from 510 to 4609 µs/cm at 25°C, against the acceptable limit of <1500 µs/cm at **Keywords:** 25°C. the concentration of Pb (lead) in the water samples varies from 0.001 to 0.011(mg/l) with an average of 0.00868±1.8 mg/l, Chromium (Cr) and Cadmium Radionuclides, Metal contamination, (Cd) concentration varies from 0.001 to 0.004 (mg/l). The result from the study **Environmental Impact** indicated that Ona River is toxic and it should not be taken as a source of drinking water without due purification. Assessment.

INTRODUCTION

The human environment is a compartment of natural ionizing radiation emission originating from the process of radioactivity. Radioactivity is a natural phenomenon and resulting radiation is ionizing and capable of impacting molecules within cells, particularly DNA molecules (Gissela et al., 2015). The risks associated with ionizing radiation must be assessed and controlled for he protection of people and the environment. Naturally occurring radionuclides (⁴⁰K, ²²⁸Ra and ²³²Th) and over 90 radioactive contaminants are present in drinking water (EPA, 2004), these contaminants are potentially hazardous, and pose threats to human health. The three main pathways the contaminants enter the human body include inhalation, ingestion and direct exposure; however, ingestion is the major pathway through which the contaminants get into the human

body. Ionizing radiation being tasteless, odorless and invisible, it is practically impossible for any human organ to either detect or be immune to ionizing radiation or can innately detect its presence. A long term exposure to a relatively large amount of ionizing radiation may result in serious health problems particularly kidney disease, impaired immune system, cancer, and anemia among other health effects that may result from ingestion of radionuclide (Turham *et al.*, 2019).

Industrialization and rapid urbanization has led to environmental pollution and exposure of our natural environment to synthetic and toxic chemicals including heavy metals. These potential elements accumulate in the environment and cause a decrease in wildlife and species of animals as a result of the toxic pollution of the ecosystem (Purushotham, 2013). Heavy metal

detrimental effects on the environmental may be elevated when they are interchanging within ecosystems through direct or indirect contact with water, soil and air. Huda et al. (2021) reported that iron, lead, cadmium and copper, nickel, cadmium iron, carbon, Chromium, silicon, molybdenum and nickel metals are used in the manufacturing of many consumer goods. These products later become wastes and some ended up in rivers, however, these heavy metals are resistant to traditional elimination procedures and do not have biodegradability (Sall *et al.*, 2020).

Safe drinking water is essential to healthy living; its availability should be a top priority for both individual and governmental/non-governmental agencies. In Nigeria, access to safe drinking water has become a great challenge with about 56 percent of the populace lacking access to a safe drinking water source (UNICEF, 2021). Only a few wealthy Nigerians who can afford to sink deep wells and boreholes drink clean and safe water while the majority drink poor and unsafe water (Alausa *et al.*, 2017).

Ona River is one of the major rivers in Ibadan, water from Ona River serves many purposes ranging from bathing, laundry, crop irrigation, livestock drinking water and a source of drinking water for the destitute. People who live far from Ibadan also take the water for daily sustenance. Ona River is poised to many contaminants resulting from the disposal of chemicals, animal wastes; pesticides; e-waste, wastes injected from the underground; and naturally-occurring radionuclides. Ibadan city is highly urbanized and its increasing population demands that the residents occupied every available space along or on the banks of the Ona River and consequently pollute the water body with waste (Jibiri, 2011). However, several studies and reports on Ona River have not included radiometric and toxic heavy metals assessment, Indeed, data on naturally occurring radionuclides and heavy metals toxicity in Ona is very sparse thus the motivation for this study. Therefore, the present study investigated levels of natural radionuclides (⁴⁰K, ²²⁸Ra, and ²³²Th) and heavy metals with their distributions and determine the radiological risk and potential health impact on the population

MATERIALS AND METHODS Geology of the Study Area

Ibadan is situated on a crystalline basement complex on Lat. 7°24' 48"N and long. 3°54' 50"E, in Southwestern Nigeria. Ibadan city spreads over undulating plains and quartzite hills characterized by moderately steep gradient hills with numerous drainage lines and soil that range from light sandy loam to sandy clay-loam. Ibadan, as the largest in West Africa has a land mass area of approximately 3080 km² with a population of 3.5 million people. The annual climatic condition interchanges between a wet season (April to October) and a dry season (November to March). The Ona River in particular lies between Lat. 3°35' and 4°10'N and Long. 7°2' and 7° 4' E. The river channels through a highly-populated area of the city, which invites indiscriminate domestic wastes into its water body. The municipal refuse and eroded soil sediments contribute to the low course stagnation of the river, in addition, to the poor color, taste and odor of the water body.



Figure 1: Map showing River Ona.

Sample Collection and Preparation

A total of fifteen water samples were randomly collected at different locations along the route of Ona River shown in Figure 1. At each sampling point, the pH meter (model: 8803 Schwerzenbach) manufactured by Mettler Toledo Group in Switzerland and the centigrade thermometer were respectively used to measure the pH and temperature of the samples immediately after collection. The pH value of each sample was taken after dipping the pH probe in the water sample for a few minutes to obtain a steady reading. 500 ml each of the samples were poured into Marinelli plastic containers after rinsing with dilute tetraoxosulphate (VI) acid (H₂SO₄) and dried to avoid contamination of the water (Mahmoud et al., 2014). The plastic containers were thereafter firmly sealed for four weeks to ensure a state of secular equilibrium between ²²⁶Ra and ²²⁸Ra and their respective gaseous progenies before gamma spectroscopy.

Determination of activity concentrations

After keeping the samples for 4 weeks to attain secular equilibrium, a 5 cm \times 5 cm solid NaI(Tl) gamma-ray

spectrometric manufactured by ORTEC and coupled to a Digital-based multi-channel analyzer (MCA) was used to count the activity concentrations of ⁴⁰K, ²²⁶Ra and ²³²Th. The detector has a poor energy resolution of about 8% at an energy of 0.662 MeV. This is considered adequate to distinguish the gamma energies of interest in the study. In addition, the photons emitted by the samples would sufficiently be discriminated if the emission probability and energy were high enough and the surrounding background continuum was low enough. However, the activity concentration of ²¹⁴Bi determined from its 1.76 MeV gamma ray peak was chosen to provide an estimate of ²²⁶Ra in the rock samples, while that of the daughter radionuclide ²⁰⁸Tl determined from its 2.61 MeV gamma ray peak was chosen as an indicator of ²³²Th. The activity concentration of ⁴⁰K was determined from 1.46 MeV Gamma rays emitted during the decay of ⁴⁰K. The standard reference sample used for efficiency calibration from Rocketdyne Laboratories was California, USA, traceable to a mixed standard gamma source (Ref No 48722-356) by Analytic Inc., Atlanta, GA, USA.

Each sample was placed on top of the well-shielded and housed detector and counted for 36,000 seconds (10h). The data acquisition, display and on-line spectrum analysis were carried out using the Genie 2000 spectroscopy software from Canberra.

Equation (1) shows the usual relationship between activity concentration (Bq/kg) and the count rate under the photo peak of a given gamma-ray spectrometry detector as expressed by Alausa et al. (2020):

$$A_c = \frac{N_c}{\varepsilon_p I_V V} \tag{1}$$

where A_c is the sample's activity concentration (Bq/kg), N_c is the net area under the corresponding peak per second, ε_p is the detector efficiency at the specific γ -ray energy I_{γ} is the absolute transition probability of the specific gamma-ray and V is the volume of the water sample in cubic meter.

An empty container of the same geometry as the sample container was counted for the same time to take care of the background radiation count and determination of the radionuclide detection limits. The detection limits (DLs) which describe the operating capability of the detector without the influence of any sample were determined using Kitto et al. (2006) model.

The detection limits (DLs) obtained in the present study were 0.12, 0.14 and 0.40 Bqkg⁻¹ for ⁴⁰K, ²²⁶Ra and ²³²Th respectively. The activity concentrations of ⁴⁰K, ²²⁶Ra and ²³²Th less than the corresponding values of the DLs are referred to as below the detection limit (BDL). One-half of each DL is considered for calculating the mean activity concentrations of the radionuclides and the radiological parameters (Alausa and Odusote, 2013)

Determination of Heavy Metal Concentrations

50 ml of the water sample was measured and transferred into a Kjeldahl flask and 20 ml of aqua regia (mixture of concentrated hydrochloric acid and nitric acid in a ratio 3:1) were added. The mixture was digested on a hot plate for 2 hours in a fume cupboard until the brown fumes disappeared. The digest was allowed to cool and filtered into a 50 ml standard flask and thereafter made up to the mark with distilled water. This was then transferred into a clean plastic bottle and analyzed for metal content using PerkinElmer AAnalyst 400 AA Spectrometer.

Effective dose due to ingestion of water

Effective doses due to ingestion of water were calculated using UNSCEAR, (2000)

 $E_d = A_c A_i C_f \tag{2}$

where E_d the effective dose (mSvy⁻¹), Ac is the activity concentration (Bql⁻¹), A_i is the consumption rate of water (l/year). According to (WHO, 2003), the dose was estimated by considering a consumption rate is 730 litre/year for adults and 512 litre/year for children. The dose conversion factors C_f were (2.8×10^{-7} , 2.3×10^{-7} , 6.2×10^{-9} for adults) and $(1.5 \times 10^{-6}, 2.5 \times 10^{-7}, 7.6 \times 10^{-9}$ Sv Bq⁻¹for children) for ²²⁶Ra, ²³²Th and ⁴⁰K, respectively (ICRP,1996; WHO, 2011).

RESULTS AND DISCUSSION

Activity concentrations of the radionuclides in water from the study area

Activity concentrations of ⁴⁰K, ²²⁶Ra, and ²³²Th (Bql⁻¹, pCil⁻¹) are presented in Table 1. The premise to which the interpretation of results in the present study is based on the safe limit recommendations and regulations of Uranium/Radium concentration ratios in drinking water by notable health and environmental protection agencies. However, the activity concentration ranges from 0.16-14.32 Bql⁻¹, (4.32-386.64) pCil⁻¹, for ⁴⁰K. The activity concentrations of ²²⁶Ra in the water samples ranged from 0.01-11.14 Bgl⁻¹, (0.27-300.7) $pCiL^{-1}$ and activity concentrations of ²³²Th ranged from 0.961-8.196 Bql⁻¹, (25.92-221.13) pCil⁻¹. From Table 1, it could be seen that samples L2, L4, L9, L12 have values within the safe limit, this means that about 30 percent of the sampling location have water sample that is fairly good for human drinking. This indicated that the water body is only being polluted by environmental factors including refuse dump by the residence, open defecation, dumping of industrial waste by nearby industries where the river cross, and throwing some electronic wastes from some popular markets where the river passes. A large percentage of results of activity concentration of natural radionuclides in the river as indicated in the present study was higher than the recommended safe limit by the World Health Organization (WHO 2006).

Annual effective dose due to ingestion of water from the study area

The total effective doses resulting from ²²⁶Ra, ^{232Th}, and ⁴⁰K radionuclides with corresponding average values for two age groups: children and adults are presented in Table 2. The annual effective dose as shown in the table ranged from 0.03 to 0.30 mSvy⁻¹ for adults and 0.75 to 4.88 mSvy⁻¹ for children. However, the mean value of an annual effective dose for adults is 0.12±0.08 mSvy⁻¹ and that for children is 2.32 ± 1.27 mSvy⁻¹ in the present study. The results of the present study were compared with the recommended values of 1.0 mSvy⁻¹ for adults and 0.20 mSvy⁻¹ for children (WHO, 2006). The mean annual effective dose result for adults in the present study is about ten times lower than the WHO standard value and the mean dose value for children is over ten times higher than the WHO standard limits. The result of the present study has shown that the annual effective dose for children is 2.20 mSvy⁻¹ higher than the dose received by adults.

annual effective dose(adults) 📕 Increase 📕 Decrease 📕 Total 2 0.3 1.8 1.6 0.12 0.05 0.21 1.4 0.04 1.2 0.2 1 0.03 0.19 0.8 0.16 0.6 0.12 0.08 0.4 0.09 0.1 0.2 0.08 0.07 0 L2 L3 L4 L5 L6 L1 L7 L8 ٤١ L15 L10 L11 L12 L13 L14 Figure 2: Annual effective dose for adults



Figure 3: Annual effective dose for children

Lifetime Cancer Risk Assessment (R)

Lifetime cancer risk assessment (R) was calculated using EPA, (1999):

$$R = D_a \times D_l \times R_f \tag{3}$$

where D_a is annual effective dose equivalent measured in Svy⁻¹, D_l is the duration of life (55.2 years for Nigerians) and R_f is the risk factor (Sv⁻¹). According to ICRP (1996), the risk assessment probability coefficient is 7.3×10^{-2} Sv⁻¹. The results of the lifetime cancer risk are presented Table 2. From the table, the results of lifetime cancer risk obtained in the present study are low when compared with the world recommendation of 8.4

 \times 10⁻³ (UNSCEAR 2016) corresponding to 2.4 mSvy⁻¹. The result indicated that no radiological cancer risk is expected from the result of the study.

Heavy metals analysis

Various analytical results have been made on the samples of water collected from the study area ranging from pH. Electrical conductivity. Total Dissolved solids (TDS) and heavy metals concentration. The pH range from 5.36 to 7.2, this is within recommended safety limit (WHO 2011). However, in terms of electrical conductivity, the value was between 884 and 4310 µs/cm at 25°C compared to the permissible limit of <1500 µs/cm at 25°C (WHO 2011). The estimated total dissolved solids (TDS) also vary from 510 to 4609 µs/cm at 25°C, against the acceptable limit of <1500 µs/cm at 25°C (WHO 2011). This may be due to municipal waste, industrial waste and e-waste constantly dumped into the water body of the river and weathering process that may be an additional factor. From Table 1, the concentration of Pb (lead) in the water samples varies from 0.001 to 0.011(mg/l) with an average concentration of 0.00868±1.8 mg/l. This mean value is enormous when compared with other literature, however it is slightly below the limit of 10 µg/l set by the World Health Organization (WHO 2011).

Chromium (Cr) concentrations vary from 0.001 to 0.004 (mg/l) as shown in Table 1, with an average concentration of 0.002063 mg/l. All the values of chromium from the sampling locations are below the recommended safety values of 50 μ g/l for non-occupational exposure. (Paul *et al.*, 2007). Human exposure to chromium is through inhalation and skin contact. However, chromium is highly toxic and

carcinogenic as reports have shown that chromium is a powerful oxidizing agent and highly soluble in water and thus dangerous. Cadmium can be fatal if inhaled or ingested and the major route of exposure to cadmium is through inhalation. Chromium (Cr), Cadmium (Cd) and lead (Pb) are not essential for human health but are considered toxic elements in nature. Meanwhile, manganese (Mn), Iron (Fe), Copper (Cu), and Zinc (Zn), are essential micronutrients for biological functions of the human body.

CONCLUSION

The activity concentration of ⁴⁰K, ²²⁶Ra and ²³²Th and heavy metals analysis in the water samples collected from Ona River has been studied. The average values obtained ⁴⁰K, ²²⁶Ra and ²³²Th were 21.89 Bgl⁻¹, 5.53 Bql⁻¹,3.87 Bql⁻¹ respectively, the results implied that the activity concentrations of ⁴⁰K, ²²⁶Ra and ²³²Th were comparable with the world standard limits and with other authors around the world. However, the calculated radiation hazards were below threshold limit values and thus the water from the study area poses no radiological health risks. In addition, the estimated total dissolved solids (TDS) was higher than the safety limit, All the values of chromium from the sampling locations are below the recommended safety values. Pb (lead) in the water samples was greater than values reported from other literature in Nigeria, however the values are slightly below the limit set by World Health Organization. Other heavy metals including manganese (Mn), Iron (Fe), Copper (Cu), and Zinc (Zn), were not found in significant amount in the water samples in the present study.

Table 1: Activity concentrations of ²²⁶Ra, ²³²Th, and ⁴⁰K in the water from Ona section of Ona River

S/n	Samples	⁴⁰ K		2.	³⁸ U		²³² Th		
		BqL ⁻¹	pCiL ⁻¹	BqL ⁻¹	pCiL ⁻¹	BqL ⁻¹	pCiL ⁻¹		
1	L1	11.33±0.5	305.91±11.4	0.91±0.7	24.57±64	3.12±0.6	84.24±16.2		
2	L2	2.17 ± 0.6	58.59 ± 1.9	0.14 ± 0.1	3.78±0.7	3.44±0.5	92.88±14.0		
3	L3	3.07 ± 0.8	82.89 ± 1.1	4.58±0.2	123.66±64	7.23±0.1	195.21±2.7		
4	L4	12.5±0.6	337.5±1.4	0.91±0.8	24.57±99	3.66±0.1	98.82±2.7		
5	L5	4.11±0.1	110.97 ± 2.0	2.29±0.2	61.83±76	1.62 ± 0.2	43.7442.7		
6	L6	7.95 ± 0.7	214.65 ± 3.1	3.43±0.7	92.61±1.4	2.95 ± 0.8	79.65±18.9		
7	L7	0.16 ± 0.01	4.32±0.4	5.91±0.4	159.57±61	2.37±0.7	63.99±12.7		
8	L8	13.7±0.7	369.9 ± 1.8	6.87±0.5	$185.49{\pm}61$	2.41±0.3	65.07±8.7		
9	L9	9.58 ± 0.5	258.66 ± 5.6	0.01 ± 0.001	0.27 ± 0.01	1.57 ± 0.2	42.39±2.7		
10	L10	14.32 ± 0.5	$386.64{\pm}1.4$	5.82 ± 0.4	$157.14{\pm}61$	4.01 ± 0.4	108.27±12.7		
11	L11	10.1±0.7	272.7±1.2	0.01 ± 0.001	0.27 ± 0.01	2.12±0.6	57.24±16.2		
12	L12	9.32±0.3	$251.64{\pm}1.8$	8.3±0.01	224.1±21	2.01 ± 0.2	54.27±4.9		
13	L13	6.41±0.5	173.07±1.6	1.52 ± 0.02	41.04±0.6	0.96 ± 0.1	25.92±2.7		
14	L14	4.36 ± 0.4	117.72 ± 1.0	4.05±0.4	109.35 ± 96	8.19±0.6	221.13±16.2		

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15 L1	5 10.15±0.9	274.05±1.8	11.14±0.7	300.78±1.6	2.17±0.6	58.59±16.2
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S/N	Effective dose (adult)	Effective Dose (children)	Lifetime cancer risk(R)x 10 ⁻³
1	0.07	2.72	0.32
2	0.08	2.65	0.25
3	0.10	4.88	0.81
4	0.09	2.81	0.36
5	0.08	1.16	0.32
6	0.12	2.29	0.48
7	0.16	1.86	0.64
8	0.19	1.90	0.77
9	0.03	1.21	0.12
10	0.20	3.13	0.81
11	0.04	1.63	0.16
12	0.21	1.60	0.85
13	0.05	0.75	0.2
14	0.12	5.03	0.89
15	0.30	1.22	0.12
Mean±6	0.12 ± 0.08	2.32±1.27	0.47±0.29

Table 2: Effective doses (mSvy⁻¹) and lifetime cancer risks due to ingestion of water from Ona section of Ona river

Table 3: Heavy metal concentration in Ona River

S/n	Sampl e code	Cd (mg/l)	Co (mg/l)	Cr (mg/l)	Cu (mg/l)	Ni (mg/l)	Pb (mg/l)	Zn (mg/l)
1	L1	0.001	ND	0.002	0.008	0.01	0.02	0.002
2	L 2	0.004	0.001	0.003	0.002	0.002	0.02	0.001
3	L 3	0.003	0.001	0.002	0.011	0.003	0.011	0.005
4	L 4	0.004	ND	0.003	0.009	0.004	0.005	0.001
5	L 5	0.001	ND	0.002	0.001	0.001	0.002	0.004
6	L 6	0.004	0.001	0.001	0.003	0.007	0.005	0.002
7	L 7	0.003	ND	0.004	0.004	0.011	0.013	0.007
8	L 8	0.001	ND	0.002	0.002	0.02	0.01	0.001
9	L 9	0.003	0.002	0.001	0.003	0.001	0.001	0.002
10	L 10	0.002	ND	0.003	0.001	0.003	0.002	0.003
11	L 11	0.004	0.001	0.001	0.003	0.007	0.005	0.002
12	L 12	0.001	ND	0.002	0.004	0.001	0.012	0.001
13	L 13	0.001	ND	0.002	0.002	0.02	0.01	0.001
14	L 14	0.003	ND	0.001	0.003	0.01	0.001	0.002
15	L15	0.001	0.001	0.002	0.008	0.01	0.02	0.002
	Mean	0.0023±1	0.0011±6	0.00206±0.3	0.00406±2.3	0.00693±4.8	0.00868±1.8	0.00243±3.8

	K	Ra	Th	Cd	Со	Cr	Cu	Ni	Pb	Zn
K	1									
Ra	906	1								
Th	602	$.845^{*}$	1							
Cd	472	$.859^{*}$	$.886^{*}$	1						
Со	231	.479	.321	.708	1					
Cr	.420	743	772	955**	828*	1				
Cu	164	331	666	731	406	.730	1			
Ni	.504	814*	796	979**	825*	.971**	.633	1		
Pb	.520	779	790	968**	808	.946**	.601	.999**	1	
Zn	756	.710	.723	.792	.654	844*	415	828*	862*	1

Table 4: Pearson correlation matrix of activity concentrations and heavy metals in Ona river

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