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DETERMINATION OF ELEMENTAL CONCENTRATION OF WATER SAMPLES USING GAMMA SPECTROSCOPY TECHNIQUE: A CASE STUDY OF TUDUN WADAN DANKADE, KANO STATE NIGERIA

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ABSTRACT
This research was aimed at investigating the Elemental Analysis of Some Metals composition of twenty (20) water samples from the different sources (Borehole, wells, Irrigation and Tap) within Tudun Wadan Dankade Local Government Area using Gamma Spectroscopy to determine the concentration of the elements present in the water sample using a well-calibrated NaI (TI) detector system. The result of the analysis was compared with the reference permissible limit to ascertain whether or not the water is safe for drinking. The mean radioactivity content obtained for K-40, U-238 and Th-232 were 7.22±0.54 Bq/L, 10.5±1.96 Bq/L and 1.71±0.16 Bq/L from Borehole water, 6.30±0.50, 4.41 ± 0.84 and 1.71 ± 0.16 in Irrigation water, while 7.53±0.62 Bq/L, 2.72±0.62 Bq/L and 3.23±0.22 Bq/L in Tap water and 15.76±1.24 Bq/L, 17.43±3.78 Bq/L in well water respectively. The mean values 5.54nGyh^{-1}, 4.52nGyh^{-1}, 2.59nGyh^{-1} and 10.08nGyh^{-1} were obtained for the absorbed dose in water sources samples of Borehole, Irrigation, Tap and well respectively while 0.67mSvy^{-1}, 0.79mSvy^{-1}, 0.60mSvy-1 and 1.93mSvy^{-1} were obtained for the Annual Effective Doses (AED). Similarly, the radium equivalent (Raq) were 13.50Bq/L, 10.16Bq/L, 5.88Bq/L and 29.59Bq/L in water for all the Borehole, Irrigation, Tap and well water, respectively. The external and internal radiation hazard indices were 0.06, 0.04, 0.02, 0.13 and 0.04, 0.03, 0.01, 0.08 respectively in water sample sources. The values obtained for the Excess Lifetime Cancer Risk (ELCR) in (x 10^{-3}) for all the water sources of Borehole, Irrigation, Tap and Well water were 2.35, 2.76, 2.10 and 6.75 respectively. It was found that the values of some exposure rate, radioactivity contents and radiological impact parameters in the study area which were higher than that of the world average values poses a serious health risk to the environment and its inhabitants.

Keywords: NaI (TI) Detector: Gamma Spectroscopy, Radiation Dose, Annual effective Dose and Radiological Impact Parameter.

INTRODUCTION

Human being are exposed to natural background radiation every day from the air, water, building materials, rocks, ground, homes, the universe and even elements in their own bodies and there is no location on Earth that is free of natural radioactivity (Kurnaz, 2013). Natural source of radiation are known to be the most significant source of public exposure to ionizing radiation (Faanu*et al,* 2012). Natural radiation sources have existed on earth, and all living organisms are continually being exposed (Saleh *et al*., 2013) and the global average annual ionizing radiation dose to person from population is 2.8mSv in total. Over 85% (2.4mSv) of this total comes from natural radiation exposure, which is mainly due to terrestrial as well as cosmic sources (Sannapa *et al,* 2016). Natural radioactivity in environmental matrix is mainly due to ^{238}U, ^{40}K, ^{232}Th and ^{226}Ra, which causes external and internal radiological hazards due to emission of gamma rays and inhalation of radon and its daughters. The natural environmental radioactivity due to gamma radiation emitted from primordial radioisotopes is one of the main external sources of radiation on earth (UNSCEAR, 1993, 2000).

Natural radiation is one of the largest contributors of external dose to the world population an assessment of gamma radiation from natural sources is of particular importance (Lee *et al,* 2009; Martin, 1972). Terrestrial radioisotopes are found on the earth that came into existence with the creation of the planet (Wicks, 2011). Some of these radioisotopes take a long time to decay and become non-radioactive (on the order of hundreds of millions of years), and are still part of the human and non-human biota today.

Natural environmental radioactivity arises mainly from primordial radionuclide such as ^{40}K, and also from ^{238}U and ^{232}Th decay series, which occur at trace levels in all ground formation (Tzortzis*et al,* 2014). It is important to monitor the terrestrial background radiation mainly due to these natural radionuclide in soil and water. Exposure to natural radioactivity depends primarily on the geographical and geological conditions, and also appears at different levels in each part of the world (UNSCEAR, 1993).

Overall exposure to natural radiation sources contributes to more than 99% of radiation dose to population (excluding medical exposure). There is only a very small worldwide contribution from nuclear power production and nuclear weapons testing (UNSCEAR, 2000).

Physical and chemical processes observed occurring following the radiation exposure involves successive changes at molecular, cellular, tissue and whole body levels that may lead to a wide range of health effects varying from simple irritation, radiation induced cancer and hereditary disorder to immediate death (ASTDR, 1999). Drinking water may contain radioactive substances that could pose a risk to human health. Naturally occurring radionuclide, in contrast

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can potentially enter water supply system at any point, or at several points prior to consumption. For this reason, naturally occurring radionuclide in water are often less amenable to control. Radiation has effects on human depending on the dose absorbed. High radiation dose may alter the DNA of human while low dose may have no appreciable effect. Biological effects of radiation exposure are classified as stochastic effect or deterministic effect (Hall, 2000). A deterministic effect has a threshold of dose, and severity of the effect is dose-related for example skin reddening while stochastic effect has no dose threshold and it is based on the molecular mechanisms involved, example of this is cancer or a heredity defect.

MATERIALS AND METHODS

The Study Area

Tudun Wada Dankade is a Local Government Area in Kano State, Nigeria. Its headquarters are in the town of Tudun Wadan Dankade, which was among the 44 Local Government Areas of Kano state Nigeria and was geographically located in the southern part of the state with a population of about 228, 658 at the 2006 census. It also shares common boundaries with Doguwa LGA, Samaila LGA, Kibiya LGA, Rano LGA, Bebeji LGA. Tudun Wada Dankade Local Government becomes one of the most extensively irrigated Local Government in the State due to the introduction of irrigation system in the area. As an agricultural town, Tudun Wada Dankade was known for the production of food stuffs and Vegetable Crops both during Dry seasons and Rainy seasons. The Dry seasons mostly start from October to April, while the Rainy season begins from April to September with an average annual rainfall of 134.4mm. The people of the local Government are 80% percent farmers who are engaged in mixed farming in both the seasons. Some of the crops produced in the area are Rice, Wheat, Maize, Millet, Guinea-corn, Beans, Tomatoes, Onions, Sugarcane, Cucumber, Cabbage, Water melon, etc.

Sample Collection

In other to measure the natural radioactivity of water sources, Twenty (20) samples four from each of Borehole water, Well water, Irrigated water and Tap water was collected from the study area at random whereas the Tap water sample marked as Tap water sample A - E (TWA-TWE). The water Samples was collected in a clean 1Litre sample collection bottles with tight covers. Irrigation water samples was collected with the aid of a bailer which was marked as Irrigation water Sample A – E (IWA-IWE) , the ground water in the wells was first purged by drawing it out severally to ensure fresh samples was obtained, marked as Well Water Sample A to E (WWA-WWE). The borehole water sample was collected after evacuating the existing water in the pipe also marked as Borehole Water Sample A- E (BWA-BWE). To avoid contamination, the water was preserved with concentrated Nitric acid to minimize precipitation and absorption of particulates in the water on containers walls. The bottle was also filled to the brim without any head space to prevent CO_2 from being trapped because it could dissolve in water and affect the chemistry. In order to achieve accuracy, sample was transferred to the Laboratory immediately after collection to be analyzed within 3 days so that the sample composition could remain intact.

The samples collected was heated on a hot plate for about 1hour and allowed to be digested up to dryness at 100^oC, allowed to cool at room temperature and filtered in to a standard 60mL sample bottle and was filled up to the mark with distilled water. The filtrate will then keep ready for analysis.

The elemental analysis will be carried out using atomic absorption spectrophotometer method and Gamma Spectroscopy method so as to achieve the accuracy level of the measurement. A different concentration of standard solutions was run on the instrument to obtain the calibration curves for each metal using measured absorbance and the corresponding concentration. The concentration selected elements was obtained from the calibration curve plotted.

All the samples (Water) were well labeled then sealed in cylindrical air tight polyvinyl chloride containers previously and thoroughly washed with dilute HNO3 and rinsed with distilled water and kept for at least 28 days so that the radionuclide in them can attain secular equilibrium after which the activity concentrations were determined on the basis of dry weight in Bq L^{-1}. After the secular equilibrium was attained, the gamma spectrometry measurements of the samples were carried out using a well calibrated Sodium Iodide (NaI(TI)) detector at the Centre for Energy Research Development (CERD).

Absorbed Dose Rate

The absorbed dose rate “D” (nGyh^{-1}) in water due to activity concentration of ^{238}U, ^{232}Th and ^{40}K was calculated using (Orosun *et al*., 2016)

$$DR = c_U Cu + c_{Th} CTh + c_K Ck \quad (1)$$

where Cu, CTh, Ck are the radioactivity concentration in Bq L^{-1} and C_U, C_{Th} and C_K are dose conversion factors for ^{238}U, ^{232}Th and ^{40}K, respectively. The values of C_U, C_{Th} and C_K used in this work are 0.462, 0.604 and 0.0417nGyh^{-1}.

The Annual Effective Dose

The annual effective dose resulting from the ingestion of water was estimated based on the assumption that a daily intake of water per person is 2L d^{-1} (WHO, 2011) from the following expression (Orosunet al., 2018; Avwiri, *et al*., 2013).

$$\text{AEDE (mSv y}^{-1}) = 365 \sum_j^j (\text{Ij x Dj}) \quad (2)$$

Where; AEDE is the annual effective dose, I_j is daily intake of radionuclide (Bq/d) = (Concentration of radionuclide in water Bq/L) x (consumption rate of water in L d^{-1}) and D_j is the ingestion dose coefficient for adults. The standard dose ingestion coefficient is equal to 0.045 μSv Bq^{-1} for ^{238}U, 0.23 μSv Bq^{-1} for ^{232}Th and 0.0062 μSv Bq^{-1} for ^{40}K (ICRP, 1994; UNSCEAR, 2000; Orosun *et al*., 2018).

Radium Equivalent Activity Index (Ra_{eq})

Radium equivalent activity index (Raeq) allows a single index or number to describe the gamma output from different mixtures of ^{238}U, ^{232}Th and ^{40}K in a material. It was calculated using the formula;

$$Ra\ eq = Cu + 1.43CTh + 0.077Ck \quad (3)$$

Where Cu, CTh, CK are the activity concentration in Bq L^{-1} of ^{238}U, ^{232}Th and ^{40}K.

Excess Lifetime Cancer Risk (Elcr)

Excess Lifetime Cancer Risk was calculated using the following equation;

$$ELCR = AEDE \times DL \times RF \quad (4)$$

Where ELCR IS the excess lifetime cancer risk, AEDE is the annual effective dose equivalent, DL is the average duration of life (estimated to 70 years) and RF is the risk factor (Sv^{-1}) that is cancer risk per Sievert. For Stochastic effect ICRP used, RF=0.5/Sv equivalent to 5.0 x 10^{-5}/mSv (UNSCEAR, 2000).

Radiation Hazard Indices

These indices are used to estimate the level of gamma radiation hazard associated with the natural radionuclide in samples. The external radiation hazard (Hext) and the internal radiation hazard (Hint) was calculated as follows:

$$H_{ext} = (CU/370) + (CTh/259) + (CK/4810) \quad (5)$$

$$H_{int} = (Cu/185) + (CTh/259) + (Ck/4810) \quad (6)$$

Where, CU, CTh and CK are the radioactivity concentration in Bqkg-1or Bq L^{-1} of ^{226}Ra, ^{232}Th, and ^{40}K respectively.

H_{int} should be less than unity for the radiation hazard to be negligible. Internal exposure to radon is very hazardous which can lead to respiratory diseases like asthma (Orosunet *et al*., 2018; Tufail, *et al.,* 2007).

RESULTS AND DISCUSSION

The activity Concentrations (Bq/L) of Natural Radionuclide K-40, U-238 and Th-236 in water samples of different sources collected from the study area (Tudun Wada Dankade LGA) are presented in Tables 1 and Table 2 respectively. So in this work, we shall be using them to estimate all the radiological impact parameters to further investigate the extent to which people living in the surrounding area of the factory are exposed.

Table 1: Sample Analysis Result of Radioactive nuclei concentration of K40, U238 and Th232

S/NO	SAMPLE CODE	K -40(Bq/ltr)	U-238(Bq/ltr)	Th-232(Bq/ltr)
1	Borehole Water Sample A (BWA)	16.50 ± 1.23	1.77 ± 0.40	0.99 ± 0.09
2	Borehole Water Sample B (BWB)	2.40 ± 0.19	12.92 ± 2.45	3.39 ± 0.29
3	Borehole Water Sample C (BWC)	6.76 ± 0.52	9.39 ± 1.10	1.23 ± 0.12
4	Borehole Water Sample D (BWD)	1.84 ± 0.18	11.29 ± 2.43	2.91 ± 0.28
5	Borehole Water Sample E (BWE)	8.6 ± 0.60	17.13 ± 3.42	0.02 ± 0.002
	Mean	**7.22 ± 0.54**	**10.5 ± 1.96**	**1.71 ± 0.16**
6	Irrigation Water Sample A (IWA)	3.08 ± 0.26	3.81 ± 0.73	2.37 ± 0.21
7	Irrigation Water Sample B (IWB)	7.81 ± 0.60	0.27 ± 0.05	5.29 ± 0.45
8	Irrigation Water Sample C (IWC)	4.89 ± 0.39	8.71 ± 1.50	0.38 ± 0.03
9	Irrigation Water Sample D (IWD)	10.79 ± 0.82	9.25 ± 1.92	6.56 ± 0.54
10	Irrigation Water Sample E (IWE)	4.95 ± 0.42	BDL	3.80 ± 0.33
	Mean	**6.30 ± 0.50**	**4.41 ± 0.84**	**3.68 ± 0.32**
11	Tap Water Sample A (TWA)	9.02 ± 0.67	BDL	0.77 ± 0.07
12	Tap Water Sample B (TWB)	19.68 ± 1.14	2.72 ± 0.62	3.58 ± 0.33
13	Tap Water Sample C (TWC)	8.73 ± 0.64	BDL	4.59 ± 0.37
14	Tap Water Sample D (TWD)	8.23 ± 0.62	BDL	4.34 ± 0.37
15	Tap Water Sample E (TWE)	0.16 ± 0.01	BDL	2.90 ± 0.27
	Mean	**7.53 ± 0.62**	**2.72 ± 0.62**	**3.23 ± 0.22**
16	Well Water Sample A (WWA)	2.62 ± 0.26	22.35 ± 5.18	1.47 ± 0.16
17	Well Water Sample B (WWB)	53.96 ± 4.25	57.57 ± 12.27	22.80 ± 1.10
18	Well Water Sample C (WWC)	10.53 ± 0.81	3.13 ± 0.64	5.36 ± 0.45
19	Well Water Sample D (WWD)	3.81 ± 0.31	0.82 ± 0.18	4.30 ± 0.37
20	Well Water Sample E (WWE)	7.87 ± 0.59	3.27 ± 0.62	4.35 ± 0.37
	Mean	**15.76 ± 1.24**	**17.43 ± 3.78**	**7.66 ± 0.49**
	Over All Mean	**9.20 ± 0.73**	**8.76 ± 1.80**	**4.07 ± 0.30**

Table 2: Absorbed dose rate, annual effective dose, radium equivalent activities, excess life cancer risk, external hazard index and internal hazard index rate for tudun wadan dankade local government

S/NO	SAMPLE CODE	D ($nGyh^{-1}$)	AED (μSvy^{-1})	AED ($mSvy^{-1}$)	Ra_{eq} (Bq/L)	ELCR (x 10^{-3})	Hex	Hin
1	BWA	2.10	299.04	0.30	4.46	1.05	0.01	0.02
2	BWB	4.91	100.45	1.00	17.95	3.50	0.05	0.08
3	BWC	5.36	545.57	0.55	11.67	1.93	0.03	0.06
4	BWD	7.05	867.79	0.87	15.59	3.05	0.04	0.07
5	BWE	8.28	605.00	0.61	17.82	2.14	0.05	0.09
	Mean	**5.54**	**483.57**	**0.67**	**13.50**	**2.35**	**0.04**	**0.06**
6	IWA	3.32	537.02	0.54	7.4 4	1.89	0.02	0.03
7	IWB	3.65	932.41	0.93	8.44	3.26	0.02	0.02
8	IWC	4.46	314.64	0.37	9.63	1.30	0.03	0.05
9	IWD	8.69	1,454.12	1.45	19.46	5.08	0.05	0.08
10	IWE	2.50	660.42	0.66	5.82	2.31	0.02	0.02
	Mean	**4.52**	**779.72**	**0.79**	**10.16**	**2.76**	**0.03**	**0.04**
11	TWA	0.84	170.11	0.17	1.80	0.59	0.01	0.01
12	TWB	4.24	779.51	0.78	9.35	2.73	0.03	0.03
13	TWC	3.14	810.17	0.81	7.24	2.84	0.02	0.02
14	TWD	2.96	765.93	0.76	6.84	2.66	0.02	0.02
15	TWE	1.76	487.63	0.48	4.16	1.68	0.01	0.01
	Mean	**2.59**	**602.67**	**0.60**	**5.88**	**2.10**	**0.01**	**0.02**
16	WWA	11.32	992.87	0.99	24.65	3.46	0.07	0.13
17	WWB	26.35	5,963.52	5.96	94.33	20.87	0.25	0.41
18	WWC	5.12	1,050.42	1.05	11.61	3.67	0.03	0.04
19	WWD	3.13	766.15	0.77	7.26	2.69	0.02	0.02
20	WWE	4.47	873.40	0.87	10.10	3.05	0.03	0.04
	Mean	**10.08**	**1,929.27**	**1.93**	**29.59**	**6.75**	**0.08**	**0.13**
	Overall Mean	**5.68**	**948.81**	**0.99**	**14.78**	**3.46**	**0.04**	**0.06**

Comparison of natural radioactivity concentration (Bq/L) in water samples of different sources at tudun wada dankade local Government Area Kano.

Radiological Impact Parameter in Water

The absorbed dose rate (nGy h^{-1}) in analyzed water samples were calculated using equation (1), with the results as presented in Table 2. The results of the annual effective dose resulting from the ingestion of water samples were estimated using equation (2) and presented in Table 2. The Radium Equivalent Activity Index (Ra_{eq}) in analyzed water samples was calculated using equation (3); presented in Table 2. The excess life time cancer risk for the analyzed water samples were calculated using equation (4); the results are shown in Table 2.

Radiation Hazard Indices: The radiation hazard indices in analyzed water samples, both the external and the internal were calculated using equation (5) and (6) respectively. The results are shown in Table 2 for radiation hazard indices.

Absorbed Dose (D)

The mean absorbed dose rate D ($nGyh^{-1}$) in analyzed water samples of different sources of borehole, irrigation, tap and well water were 5.54 $nGyh^{-1}$, 4.52 $nGyh^{-1}$, 2.59 $nGyh^{-1}$ and 10.08 $nGyh^{-1}$ in this study it is noted that the values are lower than world average value of 57 $nGyh^{-1}$ (UNSCEAR, 2000) and hence, do not pose serious health. Similarly, annual effective dose ($mSvy^{-1}$) resulting from the ingestion of water samples were estimated. In borehole water it's ranged from 0.30 $mSvy^{-1}$ - 1.00 $mSvy^{-1}$ with mean average of 0.67 $mSvy^{-1}$, in Irrigation water sample source the

AED ranged from 0.37 $mSvy^{-1}$ - 0.93 $mSvy^{-1}$ with a mean average of 0.79 $mSvy^{-1}$, in Tap water sample it was found that it's ranged from 0.17 $mSvy^{-1}$ – 0.81 $mSvy^{-1}$ with mean average of 0.60 $mSvy^{-1}$ and in well water ranged from 0.77 $mSvy^{-1}$ – 5.96 $mSvy^{-1}$ with a mean average of 1.93 $mSvy^{-1}$. Generally, it was noted that the values estimated for all the water samples in the study area were slightly lower than the world average value of 1 $mSvy^{-1}$ except for well water which is higher than the world average value because it has higher activity concentration (^{238}U and ^{232}Th) in the study area (ICRP, 1994).

The Radium Equivalent Activity Index (Raeq)

The Radium Equivalent Activity Index (Raeq) in analyzed water samples were calculated for all the water sample sources of Borehole, Irrigation, Tap and Well water mean values were; 13.50Bq L^{-1}, 10.16 Bq L^{-1}, 5.88 Bq L^{-1} and 29.59 Bq L^{-1} which is lower than the world average value of 370 Bq L^{-1} and hence pose no serious health risk (UNSCEAR, 2000).

The excess life time cancer risk

The excess life time cancer risk for the analyzed water samples were calculated with the ranged between 1.05 x 10^{-3} – 3.05 x 10^{-3} with average of 2.35 x 10^{-3} for Borehole water, 1.30 x 10^{-3} – 5.08 x 10^{-3} with mean average of 2.76 x 10^{-3} for Irrigation water, 0.59 x 10^{-3} – 2.84 x 10^{-3} with mean average of 2.10 x 10^{-3} for Tap water source sample and 2.69 x 10^{-3} – 20.87 x 10^{-3} with a mean average of 6.75 x 10^{-3} for well water sample sources. It is noted that the mean average values for all the water source samples at tudun wadan dankande is higher than the world average value of 0.2 x 10^{-3} in the environment (UNSCEAR, 2000). This implies that staying in this area with an average of 70 years without interacting with other areas in terms of feeding and shelter may pose a serious cancer risk.

Radiation Hazard Indices

Radiation Hazard Indices analyzed in water samples, both external and internal were calculated respectively. The mean external radiation hazard index (Hext) for borehole, irrigation, tap and well water were 0.04, 0.03, 0.01 and 0.04 while the mean internal radiation hazard index (Hint) of borehole, irrigation, tap and well water samples were 0.06, 0.04, 0.02 and 0.06. However, both values were found to be lower than the world average value of 1 (unity) and therefore poses no serious health risk (Beretka and Mathew, 1985).

Conclusion

The radionuclide detected in the gamma spectrometry analysis belonged to the naturally-occurring series-decay ^{238}U and ^{232}Th as well as the non-series ^{40}K. ^{238}U and ^{232}Th concentrations in water samples were found to be higher than the world average value in the study area. The estimation of most of the radiological impact parameters such as the absorbed dose rate, annual effective dose due to ingestion of water sources from Borehole, Irrigation and Tap water sources, Radium equivalent activity and radiation hazard indices were found to be lower than the world average values. However, the annual effective dose in well water samples was found to be higher than the world average value. Similarly, the excess lifetime cancer risk values in water samples in the study area were found to be higher than the world average value. This poses health hazard that may predispose the populace in the study area to serious health risk.

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