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Naturally occurring radioactive material in groundwater: potential health risk to the inhabitants at Osino in the eastern region of Ghana

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► ***Original article***

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ABSTRACT

Background: Drinking water from deep wells and boreholes is anticipated to have high concentrations of natural radioactivity from the decay of uranium, thorium, and potassium-40 isotopes. Ingestion of water containing radioactive matter for the long term dispenses potential health risks. This study sought to assess radiological quality in groundwater from selected boreholes used for domestic purposes in Osino, Ghana. ***Materials and Methods:*** The analysis was done using gamma-ray spectrometry to determine the activity of radionuclides ^{226}Ra, ^{228}Ra and 40K in fifteen water samples collected from five selected boreholes. The samples were prepared into 1 litre Marinelli beaker, firmly closed and stored for 30 days to attain secular equilibrium between the long-lived radionuclides and their short-lived offspring. ***Results:*** The mean activity concentrations for ^{226}Ra, ^{228}Ra and ^{40}K were 5.34 BqL^{-1}±1.1 BqL^{-1}, 9.21 BqL^{-1}±2.0 BqL^{-1}, and 35.64 BqL^{-1}±5.6 BqL^{-1} respectively. The estimated radiological risks for cancer mortality and morbidity for ^{226}Ra and ^{228}Ra in the drinking water samples were below the WHO set screening level of 10^{-3}. The mean hazard quotient estimated for ^{226}Ra was 9.7 µgkg^{-1}day^{-1}. The total annual committed effective dose ranged from 5.27 x 10^{-2} mSvy^{-1} to 8.25 x 10^{-2} mSvy^{-1} with a mean of 6.87 x 10^{-2} mSvy^{-1} which was within the WHO set guidance level of 0.1 mSvy^{-1}. ***Conclusion:*** The radiological quality of the water is within the individual dose criterion and may not pose a health risk. It is paramount to monitor the radiological quality of the groundwater to provide relevant information to protect public health.

INTRODUCTION

Materials that are found in the natural environment containing radioactive elements are known as Naturally Occurring Radioactive Materials (NORM). Radionuclides of concern are long-lived radionuclides Uranium-238 (^{238}U), Uranium-235 (^{235}U), and Thorium-232 (^{232}Th), Potassium-40 (^{40}K), and their decomposition progeny namely: radium, radon, polonium, bismuth, and lead. Other radionuclides which are long-lived include ^{87}Rb, and ^{115}In. NORM is widely distributed and gives rise to a natural radiation background that varies by approximately two orders of magnitude over the earth. Natural sources of radiation contribute about 87% of the natural environmental dose to the global population. The worldwide average annual effective dose from natural background radiation is 2.4 mSv [1-7]. Even though the maternal element, ^{238}U does not present harmful effects on the environment; the breath or ingestion of its offspring ^{226}Ra is established to have a high level of risk to human organs, specifically the lungs developing lung cancer [5,7]. NORM in natural materials has low concentrations, however, most human activities in which materials are extracted from the earth may increase NORM in substances, the environment, or streams of waste. The radioactivity is based on the nature of the rock and soil from which they originate, lithologic character, and the process which results in their removal and migration [8,9]. Underground water plays a key function in providing water for drinking, farming, and industrial applications. The concerns with water quality may relate to both natural and human activities with rising issues of contamination. Groundwater quality problem accounts for a great situation confronting the world [10]. Naturally occurring inorganic contaminants are radium, arsenic, nickel, cobalt, fluoride, strontium, aluminum, and manganese [11]. Radionuclide in rocks and sediment brings radioactivity into groundwater when it goes into solution, leaching point rocks or sediment without separation by any means. The natural erosion and weathering process, mobilize natural radionuclides within rocks into groundwater or surface water [12]. The concentration of natural

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radionuclides in groundwater directly correlates with the concentrations of uranium, thorium, and their decomposition progenies in soil or the bedrock [13]. The allocations of natural radionuclides are not the same therefore, knowledge of their concentration levels is needed in humans [5, 14,15]. The presence of radionuclides in groundwater causes health hazards due to internal exposure to radionuclides from the decay of the radiological elements ingested into the body. The radioactivity of groundwater is mainly connected with the existence of dissolved long-lived radium-226 (^{226}Ra) and radium-228 (^{228}Ra) isotopes. Due to radiotoxicity, especially ^{226}Ra and ^{228}Ra, the contaminant hazard is possibly unsafe to humans in low activity [16]. Radium, a chemical element, and natural radioactive isotopes bring about offspring due to uranium to lead; thorium to lead decomposition series. The limiting points for the radium isotopes in drinking water are 1 Becquerel per litre (Bq/L) or 27 Pico curie per litre (pCi/L) for ^{226}Ra and 0.1 Bq/L or 2.7 pCi/L for ^{228}Ra [17]. Radium converts into the human body as calcium. Intake of ^{226}Ra for a long period of time causes an increase in calcium in the skeleton and the possibility of bone or sinus cancer [18,19] mentioned that countries are now included in their national legislation and regulations for the control of exposure to natural sources and establishing radiological data on exposures. More importantly, [20] reported that no data on radiological hazards or degree of vulnerability to radionuclides originating from a natural point is available to lawmakers in Ghana.

The aim of this study presented in this paper was to assess the level of natural radioactivity in underground water at Osino in the Fanteakwa South District in the Eastern Region of Ghana. The emphasis of this work was to estimate the activity concentration of the radionuclides: ^{226}Ra, ^{228}Ra, and ^{40}K in groundwater for the effective committed dose per year to the inhabitants and estimate the potential risk associated with the committed effective dose. It is important to note that only monitoring and obtaining accurate data will aid in realistic decisions and policy making regarding radiological protection measures.

MATERIALS AND METHODS

Study area

Fanteakwa District is one of the twenty-one districts of the Eastern Region of Ghana with Begoro as its capital. The district is located between longitude 000°32.5' west and latitudes 06°15' north and 06°10' south. The district shares boundaries with Kwahu Afram Plains South District, northwest of Kwahu South District, south in East Akim and Atiwa District, and east in Yilo and Upper Manya Krobo District. Fanteakwa District is bordered to the north by the Afram Plains with Volta Lake in the northwest. Osino, a town in the Fanteakwa District where the study was conducted lies within longitudes W000° 29.0' and latitudes N06° 21.0'. The town is located along the Accra-Kumasi highway, between Anyinam and Bunso Junction. Farming and vocational skills are the predominant activities among the youth. The district experiences an average annual temperature of 24 ˚C with the weather usually cold throughout the year [21]. The indigenous rocks include Birimian shaping and Voltarian metamorphoses with companion rocks including Phyllis, Schist, and Granites. The hill is hooded with iron pans, bauxite, and kaolin. The rocks are gold and bauxite bearings. Rivers are seasonal and overflow their banks when the rain falls and dry up during the dry season. Rivers are the main origin of water resources mostly for household use [21]. Osino is the district capital with a total population of 5,634 [21]. Human activities such as mining, mineral sand mining, and agricultural processes may have raised the activity concentrations of natural radionuclides in the environment due to the technological transformations of nature. The vast land is used mainly for agricultural purposes. The topographical and geological nature of the area also made it possible for a proportion of the land to be used for small-scale mining activities across the southern part such as the Abompe, Osino, and Nsuapemso districts. The rocks found in the district are suitable for building and other construction purposes and are therefore exploited for residents' benefit. The district is predominantly a forest zone with few semi-savannah areas in the northern part. The typical wet-semi-deciduous forest vegetation covers about 80 percent of the total vegetation across the district. This vegetation is suitable for the cultivation of cash crops namely: cocoa, coffee, rubber, oil palm, and citrus as well as staple food crops such as plantain, cocoyam, cassava, maize, rice, and vegetables. Most of these crops are exported which helps to generate more income for farmers, the district, and the country as a whole [21].

Sample collection and preparation

Fifteen (15) composite samples of water were collected from five (5) selected boreholes in the study area figure 1, into plastic bottles and acidified immediately to prevent adhering the radionuclide to the walls of the bottles. The bottles were made full of no space. Water samples were sent to the Ghana Atomic Energy Commission Gamma Spectrometry Laboratory for analysis. The samples were prepared into a 1 liter Marinelli beaker, firmly closed and sealed with masking tape to reduce the escape of radionuclides from the water samples. The samples were stored for 30 days to acquire secular equilibrium between the long-lived radionuclides and their short-lived offspring. The sampling and its

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procedures were repeated two consecutive times.

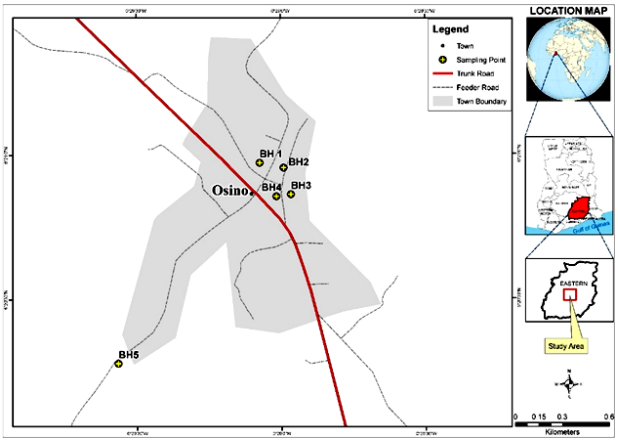

Figure 1. Groundwater sampling points in the study area.

Quality control

In sample preparation, contamination is of prime concern, and to avoid that, high-standard quality measures were taken to eliminate sample contamination during packaging, storage, and transport of samples. Marinelli beakers (IAEA standard sample container) were decontaminated using a 10% hydrochloric acid solution, rinsed with de-ionized water, and air-dried. To ensure high surface area exposure to the Gamma-ray Spectrometer, samples were acidified using 1M HCl [22] to protect radionuclides from cleaving to the walls of plastic bottles. The bottles were made full of no space to contain carbon dioxide (CO_2) gas. The detector was standardized in energy and efficiency calibration for qualitative and quantitative analysis. Other quality assurance measures taken were triplication of samples and analysis, blank preparation, and standard reference material [20]. Table 1 shows groundwater sampling points, descriptions, and their respective locations.

Table 1. Description of groundwater sampling points and location codes with their respective coordinates.

Location code	Description	Coordinates	
		Latitude	Longitude
BH 1	Market Side	N06º20.971'	W000º29.076'
BH 2	Reservoir Site	N06º20.954'	W000º28.993'
BH 3	Presbyterian Junior High School	N06º20.862'	W000º28.968'
BH 4	Mumuadu Bank (Old Site)	N06º20.855'	W000º29.018'
BH 5	Methodist Junior High School	N06º20.278'	W000º29.568'

Borehole (BH)

Instrumentation and analysis

The samples were analysed using a Gamma-ray Spectrometer consisting of a cylindrical scintillation detector (Sodium Iodide (NaI) detector) Model 3M3/3-X with a 1.2" × 1.2" transparent panel, produced by Saint-Gobain Crystals, U.S.A, in a vertical configuration. The unit is connected to Multiple-Channel Analyser (MCA) and ORTEC Maestro 32 MCB configured package to acquire a broad range of related activities, evaluations, and analyses. The detector crystal has a diameter of 63.0 mm with a length of 65.0 mm. Detector specs include: resolving power (FWHM) of 1.33MeV ^{60}Co of 1.95 keV with a proportional ratio of the output to the input of the system of 1.33MeV. The crystal is contained in an aluminium canister with a 0.5 mm thickness beryllium transparent panel entry. The maximum conversion amplification of the detector is 1024 channels. A cylindrical lead of a thickness of 20 mm with a removable cover was used as shielding to protect the detector and decrease the external radiation from the counting room or the background. The lead protection consisted of copper, cadmium, and Plexiglas of 3 mm for absorbing x-rays and photons which may be created from the lead such that the activity of the radionuclides from the drinking water at the natural level can be measured. Prior to sample analysis, the Marinelli beaker was put on the detector to acquire setting spectra of isotopes and the net peak area for correction of activity of the radioisotopes in the water. Activities of ^{226}Ra, ^{228}Ra, and ^{40}K were determined for uranium, thorium series, and potassium [20].

Energy standardisation of gamma spectrometry system

The detector was standardized for energy calibration and efficiency calibration for qualitative and quantitative analysis to be carried out. Energy calibration is required for the identification of the radioactive elements, while knowledge of the numeration ratio of output to the input of the system is needed for determining their specific activities or concentrations. Standardisations were carried out with mixed radionuclide standard, a kind of solid water, with an approximate volume of 1000 mL; 1.0 g cm^{-3} density in a 1.0 L Marinelli beaker. The mixed standard contains radionuclides that are homogenously distributed of established energies: ^{241}Am (59.54 keV), ^{109}Cd (88.03 keV), ^{57}Co (122.06 keV), ^{139}Ce (165.86 keV), ^{203}Hg (279.20 keV), ^{113}Sn (391.69 keV), ^{85}Sr (514.01 keV), ^{137}Cs (661.66 keV), ^{60}Co (1173.2 keV and 1333 keV) and ^{88}Y (898.04 keV and 1836.1 keV). Standardisation is determined experimentally in normal conditions, portraying energy dependency of the number of electrical signals in the broad range of related activities [23] using equation (1) for energy calibration [20].

$$E_n = C_0 + C_1 \times CN \quad (1)$$

Where:
E_n = energy,
C_0 and C_1 = standardisation constants of known geometry.
CN = channel number of a known radionuclide.

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Efficiency calibration

Efficiency calibration was done to acquire a broad range of related activities of the established enumeration frequency to achieve a statistical uncertainty of less than 1% with the assurance of a 95% point. The final numeration frequency estimated at the exposure maximum of the thermodynamic quantity was employed for the computation of the ratio of output to input using equation (2) to calculate the output-to-input ratio of the detector [4, 5, 24-26].

$$\eta(E) = \frac{C_T - C_B}{QE.A_{STD}.T_{STD}} \quad (2)$$

Where:
η (E) = the output and input ratio of the detector,
C_T = the total count beneath exposure maximum in the elevation array,
C_B = the background counts
Q_E = gamma-ray emission probability for the energy E,
A_{STD} = the activity of calibration well known for a known radionuclide in Becquerel (Bq) in time of measuring
T_{STD} = the counting time of standard.
The output-to-input ratio is connected to the energy equation (3).

$$(E)=A_0 + A_1 \ln E + A_2 (\ln E)^2 \quad (3)$$

Where:
ln η(E) = the output and input ratio of the detector
A_0, A_1 lnE, and A_2 $(\ln E)^2$ = calibration constants for known geometry.
Using the output-to-input ratio standardisation equation (4) the trace point is established:

$$\ln\eta(E) = 3.202 - 0.88 \ln E\_\gamma \quad (4)$$

Determination of activity concentrations

The activities of Radium-226, Radium-228, and Potassium-40 were determined in drinking water samples using the analytical equation (5); [27-30].

$$Asp = \frac{N_D e^{\lambda p T_d}}{p.T_c.\eta.m} \quad (5)$$

Where:
Asp = activity of radionuclide
N_D = final numeration radionuclide in the sample,
$e^{\lambda pTd}$ = decomposition rectification divisor of time lag during sampling and numeration
P = gamma emanation chance (gamma output),
Tc = sample enumeration period,
η = complete numeration output to input ratio of the detector scheme,
m = mass (solid) or volume (liquid) of the sample,
λp = decomposition unvarying of the mother radionuclide.

Calculation of environmental gamma dose rate

The environmental gamma dose was recorded at the sampling points with the gamma survey meter RADIAGEM 2000 manufactured by Canberra in Canada. The meter is a Geiger Muller (GM) tube for precision measurements. The survey instrument was standardised; the standardised divisor was 0.95. Values were recorded at 1 m higher up the ground of each point. The mean value obtained in microsievert per hour (μSv/h) was converted to millisievert per hour (mSv/y). The annual effective dose was estimated for the environmental gamma dose rate from the measured mean using equation (6):

$$E_\gamma, ext = D_{\gamma,ext}\, T_{exp}\, DCF_{ext} \quad (6)$$

Where:
E_γ, ext = annual effective dose
$D_{\gamma, ext}$, = outside gamma dose rate (μSv/h)
T_{exp} = vulnerability period of time in one year, 8760 hours (365 days), using outside tenancy divisor 0.2
DCF_{ext} = dosage change divisor 0.7 Sv/Gy for surrounding vulnerability to gamma irradiation [1].

Annual committed effective dose

Annual committed effective dose (H_E, ing) was measured from the activity of radionuclides in relation to the annual water ingestion rate of adults for 730 L/ year, 2 L/day x 365 days with a dosage changeover divisor for ^{228}Ra, ^{226}Ra, ^{40}K obtained out of BSS [1], using equation (7):

$$HE, \gamma_{ing}(w) = A_{sp}(w) \,.\, I(w) \,.\, \Sigma DCF_{Ing} \quad (7)$$

Where:
HE, γ_{ing} (w) = annual committed effective dose
Asp (w) = activeness of radioisotopes in a water sample in Bq/L,
I (w) = consumption of water in liters in one year,
DCF_{Ing} = consumption dose coefficient in Sv/Bq obtained out of Basic Safety Standard.

Total annual committed effective dose

The total annual committed effective dose (ET) was estimated using the dose estimation equation. An analytic equation of the total annual effective dose was set using the addition of each essential equal dose of exposure pathways regarded in this work [1] using equation (8):

Thus:
$$ET = E\gamma\,(226Ra, 228Ra, 40K) + Eing(W) \quad (8)$$

Where:
E_T = total effective dose in Sievert per year,
$E\gamma$ = environmental gamma-ray effective doses per year in the environment
E_{ing}(W) = committed effective dose ingestion of ^{226}Ra,^{228}Ra and ^{40}K in water

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Annual committed effective dose in distinct age groups of the public

The estimated annual effective dose by ingestion of ^{226}Ra was calculated because of habitual consumption of water by dwellers using equation (9):

$$HE, ing (Ra) = Cw \times CRw \times Dcw \quad (9)$$

Where:
HE, ing (Ra) = annual effective dose in ingestion of radium in water.
Cw = concentration of ^{226}Ra in consumption of water (Bq/L);
CRw = ingestion of water per year
Dcw, dose changing divisor of ^{226}Ra (SvBq/L).

Dose conversion factors of ^{226}Ra in SvBq/L for individual age groups and their respective annual water intake as suggested by [31] were used for age groups below 1 year, 1-2 years, ≥2-7 years, ≥7-12 years, ≥12-17 years and above 17 years [32].

Radiological endanger estimation of ^{226}Ra, ^{228}Ra and ^{40}K

The radiological endanger determination was to approximate the calculation of Excess Lifetime Cancer Risk (ELCR) of radioelement of drinking water samples. The ELCR could be estimated using equations (10, 32).

$$ELCR = r \times I \quad (10)$$

Where:
r = risk coefficient factor in Becquerel per litre (Bq/L);
I = per capita concentration consumption during a lifetime in Becquerel (Bq).

Mean life anticipation in Ghana is 63.4 years [33]. One year's intake of water by a person is nearly 730 L, which makes a lifetime consumption of 46,282 L. Cancer risk coefficients of ^{226}Ra are 7.17×10^{-9} Bq/L for mortality: 1.04×10^{-8} Bq/L for morbidity. Exploiting equation (10), the cancer mortality and morbidity risks for ^{226}Ra, including ^{228}Ra and ^{40}K of lifetime intake of water were estimated.

Chemical toxicity risk of ^{226}Ra

The chemical toxicity was to estimate the consequences of cancer-causing and non-cancer-causing hazards accompanied by ^{238}U in drinking water samples. The chemically poisonous risk was estimated, with a lifetime mean day-to-day dose of ^{226}Ra in water intake and a reference dose factor (RFD) of 0.6 µg/kg/day as a measure of uranium to estimate a hazard quotient. Using equations (11) and (12) respectively for the estimation of radiological hazard assessment [34-36].

$$\text{LADD for drinking water} = \frac{EPC \times IR \times EF \times ED}{AT \times BW} \quad (11)$$

$$\text{Hazard qotient} = \frac{LADD}{RFD} \quad (12)$$

Where:
LADD = lifetime mean daily dose (µgkg/day);
EPC = vulnerability to element (µg/L);
IR = water intake frequency (per day);
EF = vulnerability to element occurrences (days per year);
ED = total vulnerability to element duration (years);
AT = meantime (days);
BW = body weight (kg).

However, IR = 2 L/d; EF = 350 d/y, ED = 63.4 y, AT = 23,141 d (that is: 63.4 × 365d), BW = 70 kg (average weight of an adult).

Statistical analysis

The data were analysed using XLSTAT 2021 software for Pearson Correlation to find any association between the species and ANOVA to predict possible significant variations of the radionuclides at a 95% confidence interval level (Alpha=0.05). The data established were presented in mean and standard deviation. The sample weight and detector efficiency were taken into consideration to estimate the uncertainty using the law of error propagation. The detector background radiation takes part in the detector sensitivity among further factors that influence the minimum level of recognition of the detector. Corrections to the background radiation have been employed for the quality of the data [3, 48].

RESULTS

Activity concentrations of ^{226}Ra, ^{228}Ra and ^{40}K

The activity concentrations of the radionuclides are presented in table 2. The water samples indicated high activity concentrations in all the sampling sites. For concentrations of ^{226}Ra, ^{228}Ra and ^{40}K within the sampled locations, an analysis of variance gave p values of 0.31, 0.794 and 0.647 respectively. These were greater than their respective F values except ^{226}Ra which has F value greater than the variance p value; indicating that there was significant difference in the concentrations of ^{226}Ra, ^{228}Ra and ^{40}K (table 2).

Table 2. Activity concentrations of radium-226 (^{226}Ra), radium-228(^{228}Ra), and potassuim-40(^{40}K) in the water samples, determined by gamma-ray spectrometry.

Location Code	Activity concentrations (Bq/L)		
	^{226}Ra	^{228}Ra	^{40}K
BH 1	3.90±0.2	7.62±0.8	37.70±2.9
BH 2	5.49±0.6	11.04±0.8	31.36±2.7
BH 3	5.73±0.4	6.50±0.5	33.69±3.0
BH 4	4.65±0.4	10.29±0.7	30.93±2.7
BH 5	6.91±0.5	10.58±0.9	44.54±3.9
Mean SD	5.34±0.4	9.21±0.9	35.64±3.0
Variance of *P* value	0.31	0.794	0.647
***F*-value**	1.375	0.415	0.638

SD = standard deviation (±)

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Effective committed dose of ^{226}Ra, ^{228}Ra and ^{40}K in groundwater

The activity concentrations in table 2 were used for the estimation of the annual committed effective dose. The concentrations of individual radioisotopes of one-year committed effective dose with their respective surrounding gamma-ray for one-year effective dose, were summed up to give a total annual effective dose of groundwater (drinking water). Based on the p-value calculated above, there is significant different between the dose rates for both annual and total effective doses. Significance was predicted for the variations at a 95% confidence interval level (Alpha=0.05). Generally, the dose rates are below the global effective dose of 0.1 mSv/y (table 3).

Table 3. Annual and total annual committed effective dose due to Radium-226 (^{226}Ra), Radium-228 (^{228}Ra), and Potassuim -40 (^{40}K) in groundwater calculated for all the water sampling points analysed in this work.

Location Code	Annual Effective Dose (mSv/y)			Total Annual Effective Dose (mSv/y)
	^{226}Ra	^{228}Ra	^{40}K	
BH 1	7.97E-04	3.73E-03	1.71E-04	5.71E-02
BH 2	1.12E-03	5.40E-03	1.42E-04	7.07E-02
BH 3	1.17E-03	3.18E-03	1.52E-04	8.02E-02
BH 4	9.50E-04	5.03E-03	1.40E-04	5.27E-02
BH 5	1.41E-03	5.17E-03	2.02E-04	8.25E-02
Mean	1.09E-03	4.50E-03	1.61E-04	6.86E-02

Mortality and morbidity risk of 226Ra, 228Ra and 40K in the water samples

As presented in table 4, risks were high even though generally; mortality and morbidity values were in the World Health Organisation (WHO) set screening level of 10-3 except the 228Ra values of mortality and morbidity for BH2 and BH5, which were above the WHO screening value. P-value predicted significant differences in both mortality and morbidity between the various radionuclides, a significant variation was also predicted at a 95% confidence interval level (Alpha=0.05. The risks were within the WHO screening level.

Table 4. Lifetime cancer mortality and morbidity risk due to Radium-226 (^{226}Ra), Radium-228 (^{228}Ra), and Potassuim-40 (^{40}K) in groundwater calculated for all the water sampling points analysed in this work.

Code	Lifetime cancer mortality and morbidity risk					
	$^{226}Ra_a$	$^{226}Ra_b$	$^{228}Ra_a$	$^{228}Ra_b$	$^{40}K_a$	$^{40}K_b$
BH 1	1.29E-03	1.88E-03	7.05E-03	9.91E-03	6.16E-04	9.56E-04
BH 2	1.82E-03	2.64E-03	1.02E-02	1.44E-02	6.24E-04	9.70E-04
BH 3	1.90E-03	2.76E-03	6.02E-03	8.45E-03	6.70E-04	1.38E-03
BH 4	1.54E-03	2.24E-03	9.52E-03	8.45E-03	6.16E-04	9.56E-04
BH 5	2.29E-03	3.33E-03	9.79E-03	1.38E-02	8.86E-04	1.38E-03
Mean	1.77E-03	1.97E-03	8.52E-03	1.10E-02	6.82E-04	1.13E-03

$_a$ Mortality, $_b$ Morbidity

Mass concentration of ^{226}Ra in groundwater sample

The activity concentration of 226Ra was used to estimate the mass concentrations in microgram per litre (µg/L). P-value predicted significant difference in the data sample from all the locations. Mass concentrations were high in all the water samples, especially for samples from the location code BH 5 (table 5).

Table 5. Activity and mass concentrations of Radium-226 (^{226}Ra) in groundwater estimated for all the water sampling points analysed in this work.

Location Code	Activity and mass concentrations		
	^{226}Ra (Bq/L)	^{226}Ra (pCi/L)	^{226}Ra (µg/L)
BH 1	3.90±0.2	105.30±9.2	155.8±11.9
BH 2	5.49±0.6	148.23±10.6	219.3±13.6
BH 3	5.73±0.4	154.71±11.8	228.9±13.8
BH 4	4.65±0.4	125.55±10.9	185.8±12.7
BH 5	6.91±0.5	186.57±12.7	276.1±14.9
Mean ±SD	5.34±0.4	144.07±11.0	213.2±13.4

Estimated lifetime average daily dose and hazard quotient of radium-226

In table 6, the estimated Lifetime Average Daily Dose (LADD) from the water samples was used to estimate the hazard quotient for Radium-226. Significant difference was predicted in the data samples by the p-value, but no significance was predicted at 95% confidence interval level (Alpha=0.05) for Radium-226. Nevertheless, the hazard quotient was much greater than the global screening level of 1.

Table 6. Lifetime average daily dose (LADD) and hazard quotient of Radium-226 (^{226}Ra) estimated for all the water sampling points analysed in this work.

Code	Estimated LADD (µg/kg/day)	Hazard Quotient
BH 1	4.26±0.2	7.1±0.5
BH 2	6.01±0.4	10.0±0.8
BH 3	6.27±0.4	10.4±0.8
BH 4	5.09±0.3	8.4±0.6
BH 5	7.56±0.5	12.6±0.8
Mean±SD	5.84±0.04	9.7±0.04

Effective dose of ^{226}Ra per year for individual age groups

The annual effective dose of ^{226}Ra was relatively low in the groundwater samples from all the sampling sites for the individual age groups (table 7). Significance difference was predicted among the individual age groups by the p-value. Significance was also predicted for the variations at a 95% confidence interval level (Alpha=0.05). However, the dose rates among the different age groups were within the acceptable value of 0.1 mSv/y [38].

Table 6. Lifetime average daily dose (LADD) and hazard quotient of Radium-226 (^{226}Ra) estimated for all the water sampling points analysed in this work.

Code	Bq/L	< 1	1-2	2-7	7-12	12-17	> 17
BH1	3.90	6.69E-03	1.60E-03	1.21E-03	2.23E-03	5.76E-03	7.97E-04
BH2	5.49	9.42E-03	2.25E-03	1.70E-03	3.14E-03	8.11E-03	1.12E-03
BH3	5.73	9.83E-03	2.35E-03	1.78E-03	3.28E-03	8.47E-03	1.17E-03
BH4	4.65	7.98E-03	1.91E-03	1.44E-03	2.66E-03	6.87E-03	9.50E-04
BH5	6.91	1.19E-02	2.83E-03	2.14E-03	3.95E-03	1.02E-02	1.41E-03
Mean	5.34	7.02E-03	2.19E-03	1.65E-03	3.05E-03	7.88E-03	1.09E-03

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Correlation analysis

Pearson correlation matrix was carried out to check the mutual relationship between radionuclides in the water samples as presented in table 8. The correlation shows positive and a strong association between ^{226}Ra, ^{228}Ra and ^{40}K in the water sample.

Table 8. Pearson correlation matrix between ^{226}Ra, ^{228}Ra and ^{40}K in groundwater samples.

	^{226}Ra	^{228}Ra	^{40}K
^{226}Ra	1	0.292	0.824
^{228}Ra	0.292	1	0.446
^{40}K	0.824	0.446	1

DISCUSSION

Potassium is an important element in the human body and is rarely determined in drinking water in quantities which can affect human health [38]. The high activity concentrations of ^{226}Ra and ^{228}Ra in this work may be attributed to geological considerations. According to [39], the high activity of radionuclides is usually determined by geological substances such as eruptive rocks and ore. The location of a radioisotope is based on the location of rocks, lithological character, and the procedure by which they are removed from the soil and transported [8, 9]. According to [40], radionuclides in nature decay in stable conditions when the activities of radionuclides are the same. The activity concentrations for ^{226}Ra and ^{228}Ra were above the WHO-recommended concentrations of 1 Bq/L and 0.1 Bq/L respectively.

The borehole water BH2, BH3, and BH5 showed high activity in one year of committed effective dose for ^{226}Ra. ^{228}Ra also showed high activity concentration in one year of committed effective dose in all the samples. Potassium generally indicated a low committed effective dose; committed effective doses per year were generally low in all the samples. The estimated one-year committed effective dose of radionuclides including potassium in the drinking water was 5.75×10^{-3} mSv/y. This value is the WHO-acceptable threshold of 0.1 mSv/y. The total annual committed effective dose with a mean of 6.87×10^{-2} mSv/y was also within the WHO threshold of 0.1 mSv/y. There was a general indication of a low annual committed dose of radionuclides in all the sampling points when compared with the recommended value. According to [41], radium accumulates in bone from natural radiation if taken in drinking water. The estimated committed effective dose of members less than 1 year to 17 years and above have their respective means at 9.15×10^{-3} mSv/y for ages up to 1 year; 2.19×10^{-3} mSv/y for ages above 1 to 2 years; 1.65×10^{-3} mSv/y for age above 2 to 7 years; 3.05×10^{-3} mSv/y for age above 7 to 12 years; 7.88×10^{-3} mSv/y for age above 12 to 17 years and 1.09×10^{-3} mSv/y for age above 17 years as shown in table 7. The committed effective doses in all the age groups were within the WHO threshold of 0.1 mSv/y.

A radiological assessment was carried out because of the intake of radioisotopes in drinking water. Generally, all five boreholes recorded cancer mortality and morbidity risk within the acceptable limit of 10^{-3} [1]. Activities in uranium, and thorium increased in localized areas because of human activities or practices which caused variation in exposures [1]. The cancer mortality and morbidity risks in this study were within the WHO acceptable value of 10^{-3}. The cancer risk at 10^{-4} was low when compared to the WHO recommended limit of 10^{-3}. Several health environmental protection organizations set acceptable limitations for uranium in the intake of water by a human [42] set 15 µg/L for uranium as the acceptable value in water, [43] recommend 20 µg/L for uranium as the acceptable limit in water, and the [44], which modulates uranium in providing public water, established a 30 µg/L limit.

The mass concentration of ^{226}Ra with a mean of 213.2 µg/L in this study was far above the safe recommended values set by the various organizations. The estimated mass concentration was also high when compared to values obtained from work done in other countries as in table 9, [34].

Table 9. Comparison of mass concentration of ^{226}Ra obtained from this work with others published in literature in other countries.

Country	Mass concentration (µg/L)	References
Ghana	213.2	This work
Nigeria	157.2	[34]
Slovenia	0.5	[34]
Germany	1.15	[34]
China	1.4	[34]
Finland	1.6	[34]
Brazil	0.08	[34]

According to [42], high concentrations of uranium of more than 15 µg/L in drinkable water may exhibit harmful biological effects in humans. These differences may be a result of the Uranus state that existed in the layer yielding groundwater which might have undergone a chemical reaction. The effect of the movement of the earth's crust on fallible zones could result in the disintegration of rock minerals in the groundwater table channel [34].

Water samples from the boreholes had a relatively high estimation LADD and hazard quotient. The LADD of ^{226}Ra was at a mean of 5.84 µg/kg/day. The hazard quotients were observed in the range from 7.1 to 12.6 with a mean of 9.7. Generally, the hazard quotient values were high and above unity. The hazard quotient is a proportion of likely vulnerability to elements with a degree of no contrary effect is likely to happen. A hazard quotient ≤ 1 indicates that no adverse health effect may occur due to vulnerability to elements. A hazard quotient ≥ means contrary health consequences may happen. However, it is necessary to note that a hazard

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quotient of more than one does not necessarily mean health effects may occur [35]. Though the geochemistry of the study area may contribute to high LADD values, [45] noted that uranium is worldwide known for its radiological hazard and its chemical toxicity should be regarded [46] also observed that uranium is a heavy metal that is not responsible for specific harm; its decay products pose threat to human health and the environment. 90 µg of uranium is contained in the human body from the usual consumption of water, food, and air; 66% is contained in bone; 16% in the liver; 8% in kidneys; 10% in other tissues. ^{226}Ra is a product of ^{238}U which is more soluble in water than ^{228}Ra [47].

CONCLUSION

This study investigated groundwater mainly for its committed effective dose, radiological risk, and chemical toxicity. The total annual committed effective dose was within the WHO-acceptable threshold of 0.1 mSv/y for safe drinking water. Radiological risks of cancer mortality and morbidity were within the acceptable limit of 10^{-3}. *The estimated doses obtained may have no observable health risk.*

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***Author's contributions*:** AEM and GET designed the study. AR and GET performed the statistical analysis and wrote the first draft of the manuscript. KDO and AOK Technical design of the analyses of the study. SA and HJ reviewed the literature searchess and finalized analyses of the study. AEM and KR performed the lab work, wrote the protocol. QE mapped the site locations.

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