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Geological interactions and radio-chemical risks of primordial radionuclides ^{40}K, ^{226}Ra, and ^{232}Th in soil and groundwater from potential radioactive waste disposal site in Ghana

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Abstract
This study assessed the influence of geological depth and formation on activity concentrations of ^{232}Th, ^{226}Ra and ^{40}K in soil and groundwater, and radio-chemical health risks. Preference ranking organisation method for enrichment evaluation and geometrical analysis for interactive aid indicated significant correlation between ^{232}Th and ^{40}K, thus indicating potential similar primordial origin. Deepest depth rocks at 145 m and 148 m constituted of dark coloured silicified schist exhibited minimal activity with potential hydro-geological interactions with groundwater. Age-dependent annual effective ingestion dose for ^{226}Ra and ^{232}Th are higher than the UNSCEAR reference dose, thus the groundwater is unsuitable for long-term consumption.

Keywords Natural radioactivity · Radio-chemical risk · Multi-criteria decision making · Gamma spectrometry · Annual gonadal equivalent dose · Annual effective ingestion dose

Introduction

Natural sources of radiation also known as background radiation are naturally occurring radionuclides that exist in the environment. These natural sources involve primordial or terrestrial (such as rock and soil), cosmic radiation (such as stars and sun), naturally occurring radioactive material (NORM) emanating from the processing of raw materials (such as feedstock, final products, intermediate- and co-products, and waste), and other radionuclides of natural origin resulting from the processing of minerals in exception of nuclear installation related radioactive material and waste [1]. In addition to the natural sources anthropogenic based natural radionuclides may also be released into the environment from oil and gas processing, combustion of coal, nuclear installation related radioactive material and waste, and operation of nuclear power plants [2]. According to the United Nations Scientific Committee on Effects of Atomic Radiation (UNSCEAR), the global average annual effective dose per individual from all sources of radiation in the environment is about 3.0 mSvy^{-1}, and 80% of the radiation dose (i.e. 2.4 mSvy^{-1}) can be attributed to naturally occurring sources of radiation [3]. The natural emission of gamma radiation is due largely to primordial radionuclides [2].

Uranium and thorium series, and their progenies, as well as potassium-40 (^{40}K) which exist at trace levels in the earth's crust are the main natural primordial or terrestrial radionuclides [2]. The radionuclides concentration in soil and rocks depend on the local geology and geographical conditions, and in particular, the rock types and the geochemical processes operating in each region across the world [3]. The intensity of primordial radioactivity may vary by an order of magnitude due to the differences in geological factors across regions [4]. Uranium and thorium are fused into late crystalline magmas and residual solutions due to their large ionic radii thereby minimizing their migration from the crystalline silicate. In this regard, higher activity concentrations of uranium and thorium are found mostly in igneous rocks that include intrusive materials such as

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granites and pegmatites [4, 5]. However, some shales and phosphate rocks have comparatively high content of uranium and thorium [3].

It is reported that radionuclides can migrate from the lithosphere to other natural environments via numerous pathways mainly by erosion and weathering of terrestrial rock under the influence of anthropogenic and natural activities. The migration of these radionuclides in the environment is aided by gravity, water, and wind [2, 6]. Some of the radionuclides may end up in groundwater. The activity concentration of natural radionuclides in groundwater is associated with the activity concentrations of thorium (^{232}Th) and uranium (^{238}U and ^{235}U), and their progenies in soil and bedrock [4]. Their abundance in water is a function of their half-lives, the concentrations of ^{238}U and ^{232}Th in the sediments, and the geochemistry of the parent isotopes. It is therefore critical to examine the potential baseline interconnection of radionuclides between the geosphere and groundwater prior to the geological disposal of conditioned radioactive waste. This will help to mitigate the potential contamination of groundwater resources with radionuclides since groundwater serves as a critical source of potable water for most inhabitants, especially in the developing world [7].

Ingestion of radionuclides through drinking water and food consumption contributes significantly to the average radiation doses of various organs in the body. In this context, ingestion constitutes an essential pathway for long-term health risk assessment with respect to radionuclides exposure [3]. Drinking water and food contribute approximately 85% and 15%, respectively to ingested radionuclides [8]. The gastrointestinal absorption of radium from drinking water and food was estimated to range from 15 to 21% by ICRP [9]. As part of the exploratory investigation of any geological area for future disposal of radioactive wastes, it is relevant to establish the baseline radiological risk of the potential sites.

Geological repositories have been identified by the nuclear industry globally as one of the most viable long-term approaches in the disposal of radioactive waste. In particular, the borehole disposal system (BDS) for radioactive waste is highly pertinent due to its immense cost-benefits and minimal footprint [10–13]. In Ghana, the quest to dispose radioactive waste safely and securely in an engineered geological repository is no different, as this will help protect the health of both the public and the environment. In this regard, the Ghana Atomic Energy Commission (GAEC) has embarked upon the disposal of radioactive waste using the BDS approach. In line with the IAEA's safety protocol for the BDS, potential sites need to be characterized to facilitate well-informed site selection process to guarantee the sustainable protection of the environment and human health. The site characterization process entails geological, geophysical, geochemical, and radio-chemical investigations to establish underlining conditions for the overall safety assessment of the disposal site.

An in-depth knowledge on the activity concentrations of natural radionuclides is imperative to provide prior data on the source, dynamics, and environmental fate of radionuclides, as well as for radiological risk assessment. Although current literature is inundated with radionuclide distribution and radiological risk across various environmental matrices such as soil, water and atmospheric particles, to the best of our knowledge there is limited research information on the influence of geological depth on radionuclide distribution and radiological risk. Therefore, this study is aimed at assessing the geological interactions between the principal terrestrial radionuclides in soil and groundwater from the sites earmarked for the borehole disposal of radioactive waste using relevant multivariate and multi-criteria decision making statistical approaches. Secondly, the potential radio-chemical risks associated with the primordial radionuclides in the study area will be examined. The outcomes of this study are relevant to regulatory stakeholders in Ghana as well as the international community notably IAEA, and the findings can be extended to similar borehole disposal concepts of radioactive waste in other countries.

Materials and methods

Study area and geology of the site

The study area is a potential site for the borehole disposal of disused sealed radioactive sources, and is situated in the vicinity of the Ghana Atomic Energy Commission's (GAEC) Research Reactor and radioactive waste management facility at Kwabenya, a suburb of Accra, Ghana. The area lies within latitude 5° 6′7″ N to 5° 6′9″ N and longitude 0° 21′ W to 0° 26′ W, at an elevation of 64 m. The names of the sampling points with their respective global positions are shown in Table S1. It is situated to the north of Ghana's capital city, Accra and also to the north-west of Ghana's premier university, the University of Ghana, Legon. The study area is therefore surrounded by catchment areas with significant socio-economic activities. As a result, the area merits an in-depth characterization prior to the geological disposal of radioactive waste to facilitate sustainable environmental protection. The map of the study area with the sampling sites is shown in the Supplementary Information, Fig. S1.

The GAEC site is generally covered by a layer of reddish, pisolithic lateritic materials also called ironstone gravel. The potential disposal site for the radioactive waste is underlain by interleave Dahomenyan and Togo geological formations which are among the Precambrian metamorphic rocks in Ghana. The Togo geological formations are Upper Precambrian while the Dahomeyan formations are Middle

to Late Precambrian [14, 15]. The Dahomenyan geological formation is predominantly made-up of quartzo-feldspathic gneiss hornblende and biotite gneisses, migmatites, granulites, schists, some of which are rich in tiny marble [16]. The lithology of the three focal investigatory boreholes, BH1, BH2 and BF observed during the rotary percussive drilling process as shown in Table S2 indicates that a thin layer of mottled clay (about 1−2 m) mixed with fabrics of quartzite occur below the layer of laterite.

Sample collection and preparation

Seventeen (17) soil and rock chipping samples were collected from three (3) boreholes drilled at the GAEC Research Reactor site. A total of fourteen (14) groundwater samples were collected from ten (10) boreholes situated at various points on and around the GAEC site, and four (4) boreholes outside the vicinity of the GAEC site.

A spade full of drilling chips were collected at regular intervals and then used to identify the rock's composition (lithology) after which the samples were bagged in plain polythene zip locked bags and labelled appropriately taking into consideration the depth (in meters) at which the sample was collected. The samples were air dried in trays for 5 days and then oven dried to constant weight. The rock samples were then grinded into fine powder using a ball mill grinder to ensure homogeneity. The powdered samples were sieved through a 250 µm pore size mesh to form composite samples and then prepared into one (1) litre Marinelli beakers for analyses. The Marinelli beakers with the soil samples were sealed with paper tapes to prevent the escape of gaseous radionuclides in the samples and then stored for 30 days before measurement. The storing period was to ensure the attainment of secular equilibrium between the long-lived parent and short-lived daughter radionuclides.

The groundwater samples were collected after purging the boreholes to remove any stagnant water thereby facilitating the collection of representative samples from the aquifer system. The water samples were collected into previously clean five litre (5L) polyethylene gallons, acidified with 1 M HCl to prevent the radionuclides from adhering to the sides of the container. The gallons were filled to the brim without any head space to prevent trapping of carbon dioxide (CO_2) gas. The containers were then sealed tightly and labelled appropriately.

The groundwater samples were transported to the laboratory and then prepared into a one (1) litre Marinelli beaker, firmly closed, labelled, and sealed with masking tape to reduce escape of radionuclides. Similarly, the groundwater samples were stored for 30 days to establish secular equilibrium between the long-lived parent and their short-lived daughter radionuclides before analysis. Both soil and groundwater samples were analyzed using gamma spectrometry as described below to determine the radioactivity concentrations of the terrestrial radionuclides.

Radioactivity concentration measurement

The activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K in the samples were measured using a high resolution hyper pure germanium (HPGe) detector with a relative efficiency of 30% relative to a 3″ × 3″ NaI (Tl) scintillator. The energy resolution (FWHM) of the detector was 2 keV at 1.332 meV of a ^{60}Co source. The detector was placed in a Lead shield to reduce the background radiation originating from the building materials and surrounding cosmic rays. Advanced multi-channel analyzer (MCA) emulation software (Genie) was used for data acquisition, storage, display, and analysis of the acquired gamma-spectra. Each sample was placed on the shielded HPGe detector and measured for an accumulating time of about 16 h. Before measurement of the samples, the gamma background radiation in the laboratory was determined with an empty cylindrical container of similar geometry as the sample containers under identical measurement conditions. The measured background activity was deducted from the measured activity of each sample. In order to determine the uncertainty of activity concentration of radionuclides, a statistical formula, *S1* in the Supplementary Information which combines the uncertainties estimated during the measurement of mass or volume of the sample (µm), detection efficiency (µε), emission probability (µP) and net peak count for the sample (μN_c) was used.

Energy and efficiency calibrations were carried out with a mixed radionuclide calibration standard in the form of solid water, with an approximate volume of 1000 mL and density of 1.0 g cm^{-3} in a 1.0 L Marinelli beaker. The standard was obtained from the International Atomic Energy Agency (IAEA). The activity concentration of ^{40}K was measured directly by its own gamma ray of 1461 keV. The activity concentration of ^{226}Ra was assessed on the basis of average activity concentration of its short-lived daughter products; ^{214}Pb and ^{214}Bi at 295.21 keV, 351.92 keV and 609.92 keV, 1120.28 keV, 1764.49 keV, respectively. The decay products used for measuring the activity concentration of ^{232}Th were; ^{228}Ac at 338, 463, 911 and 968 keV and ^{212}Bi at 727 keV, ^{212}Pb at 238 keV and ^{234} Pa at 1001 keV. The activity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K in Bq kg^{-1} for the soil and Bq L^{-1} for the water samples were determined using Eq. 1 [17, 18].

$$A_{sk} = \frac{(N_A \cdot \exp(\lambda_p \cdot t_d)}{\varepsilon(E) \times p_r \times T_c \times m} \quad (1)$$

where N_A refers to the net counts of the radionuclide in the samples, t_d is the delay time between sampling and counting, $P\gamma$ is the gamma ray emission probability (gamma ray

yield), ε(E) is the absolute counting efficiency of the detector system, T_c is the sample counting time, m is the mass of the sample (kg) or volume of sample (L), $\exp(\lambda_p \cdot t_d)$ is the decay correction factor for delay between time of sampling and counting, and λ_p is the decay constant of the parent radionuclide [19].

Radiological and chemical risk assessment

The radiological and chemical (radio-chemical) risks posed to humans as a result of exposure to terrestrial radionuclides in soil and groundwater was assessed to establish the baseline risk data prior to the implementation of geological disposal of conditioned radioactive waste in Ghana. A number of relevant risk parameters were employed as elaborated below based on existing literature.

Absorbed dose rate and annual effective dose rate measurement

The absorbed dose rate (AD) was calculated using Eq. 2 below:

$$\mathrm{AD(nGy/h)} = 0.427C_{Ra} + 0.662C_{Th} + 0.043C_K \quad (2)$$

where C_{Ra}, C_{Th} and C_K are the activity concentrations of Ra, Th and K, respectively. The annual effective dose equivalent (AEDE) was estimated from the absorbed gamma dose rate using the dose conversion coefficient from absorbed dose in air to the effective dose (0.7 Sv Gy^{-1}) and the outdoor occupancy factor (0.2). Thus, the AEDE was calculated using Eq. 3 below:

$$\mathrm{AEDE(mSv/y)} = DTfQ\varepsilon \quad (3)$$

where D is the absorbed dose rate in air (nGy h^{-1}), T is the time in seconds in a year (8760), f is the occupancy factor which modifies the average time spent outdoors in the sites (0.2), Q is the effective dose rate quotient and absorbed dose rate in air (0.7 Sv Gy^{-1}), Ɛ is the factor converting nano (10^{-9}) into milli (10^{-3}). The absorbed dose rate in air is contingent on the concentrations of the radionuclides in rock and soil. In this regard, the absorbed gamma dose rates in air at one (1) meter above the ground surface (D) were estimated from the results of the activity concentration measurements of the radionuclides using Eq. 4 as proposed by UNSCEAR [3]:

$$D = \sum_x A_x \times C_X \quad (4)$$

where A_x (Bq kg^{-1}) is the activity of the radionuclides and C_x (nGy h^{-1} per Bq kg^{-1}) is the corresponding dose conversion factors. The dose conversion factors reported by UNSCEAR [3] assumed that contribution from other naturally occurring radionuclides is insignificant.

Radium equivalent activity (Ra_{eq})

Due to the non-uniform distribution of radionuclides in soil and bedrock, standardization with respect to exposure to radiation has been defined in terms of radium equivalent activity (Ra_{eq}) in Bq Kg^{-1} to compare the specific activity of materials covering different amounts of ^{226}Ra, ^{232}Th and ^{40}K [20]. Ra_{eq} was evaluated using Eq. 5 below as adapted from Uosif et al. [21], Gonzalez-Fernandez et al. [22] based on the estimation that 10 Bq kg^{-1} of ^{226}Ra, 7 Bq kg^{-1} of ^{232}Th and 130 Bq kg^{-1} of ^{40}K produced the same gamma dose rate.

$$Ra_{eq} = C_{Ra} + 1.43C_{Th} + 0.07C_K \quad (5)$$

where C_{Ra}, C_{Th} and C_K are the concentrations of ^{226}Ra, ^{232}Th and ^{40}K in Bq kg^{-1}, respectively.

Activity Utilization Index (AUI)

The activity utilization index (AUI) is usually estimated to determine the dose rate in air from different combinations of the various radionuclides present in the bedrock. The formula for the calculation is stated in Eq. 6 below [23, 24].

$$\mathrm{AUI} = \left(\frac{C_{Ra}}{50\ \mathrm{Bq/kg}}\right) * f_{Ra} + \left(\frac{C_{Th}}{50\ \mathrm{Bq/kg}}\right) * f_{Th} + \left(\frac{C_K}{500\ \mathrm{Bq/kg}}\right) * f_K \quad (6)$$

where C_{Ra}, C_{Th} and C_K refer to the concentrations of ^{226}Ra, ^{232}Th and ^{40}K in Bq kg^{-1}, respectively, and f_{Ra} (0.462), f_{Th} (0.604) and f_K(0.041) are the fractional contributions to the total dose rate in air due to gamma radiation from the actual concentrations of these radionuclides. The typical activities per unit mass of ^{226}Ra,^{232}Th and ^{40}K in soil are 50, 50 and 500 Bq kg^{-1}, respectively [25].

Annual gonadal equivalent dose

The United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR) considers certain organs with rapidly dividing cells such as gonads, the active bone marrow and bone surface cells of prime interest [3]. The annual gonadal equivalent dose (AGED) is a measure of the genetic significance of the yearly dose received by the population's reproductive organs [23]. The AGED (μSv y^{-1}) in this study was calculated using Eq. (7) below [23, 26, 27].

$$\mathrm{AGED}\ (\mu\mathrm{Sv\ y^{-1}}) = 3.09A_{Ra} + 4.18A_{Th} + 0.31A_K \quad (7)$$

where A_{Ra}, A_{Th} and A_K are the activity concentration of ^{226}Ra, ^{232}Th and ^{40}K, respectively. According to UNSCEAR [3], the average AGED value for the world is 300 μSv y^{-1}.

Excess lifetime cancer risk (ELCR)

The excess lifetime cancer risk (ELCR) gives an indication of the additional risk associated with excessive exposure to radiation and other cancer-causing agents that could lead to cancer in an individual. The ELCR can be calculated using Eq. (8) below [3, 18, 28].

$$\mathrm{ELCR}\left(\mathrm{mSv\ y^{-1}}\right) = \mathrm{AEDE} \times \mathrm{DL} \times \mathrm{RF} \tag{8}$$

where AEDE is Annual Effective Dose Equivalent, DL is the duration of life and RF is the risk factor (that is fatal cancer risk per Sievert). According to ICRP [29], the detriment-adjusted nominal cancer risk coefficients for stochastic effects after exposure to radiation at low dose rate is 0.055 $\mathrm{Sv^{-1}}$ for the whole population.

Annual effective ingestion dose

The Annual Effective Ingestion Dose (AEID) due to ingestion of the groundwater was estimated to assess the influence of the radionuclides to public exposure from natural radioactivity. The AEID (μSv $\mathrm{y^{-1}}$) was calculated as the product of activity concentration of the radionuclides in the water, the annual intake of the water, and the dose conversion factor using Eq. (9) below [30].

$$\mathrm{AEID} = A_w \times I_A \times E \tag{9}$$

where A_w (Bq $\mathrm{L^{-1}}$) is the natural radioactivity in water, I_A (L $\mathrm{y^{-1}}$) is the annual intake of water and E (Sv $\mathrm{Bq^{-1}}$) is ingested effective dose conversion factor for natural radioactivity. The value of the dose conversion factor varies for different radionuclides as well as the age of the individual. According to the World Health Organization (WHO), the annual per capita consumption of groundwater by infants, children and adults is assumed to be 250, 350 and 730 L $\mathrm{y^{-1}}$, respectively [31]. The recommended age-dependent dose conversion factors (mSv $\mathrm{Bq^{-1}}$) by WHO and ICRP are presented in Table S3 of the Supplementary Information.

Health hazard of radium

Radium toxicity due to water ingestion presents two-fold challenges. The radium element emits radiation of high ionizing power. Secondly, radium as a heavy element is associated with chemical toxicity [32, 33]. By estimation, the most radiotoxic radionuclide is radium because about 20% of ingested radium is absorbed into the bloodstream and then dispersed to bones and soft tissues. However, it is mainly retained in growing bones [34, 35].

In this study, the radiological toxicity was estimated as the life-time cancer risk (LCR) due to ingestion of radium based on the activity concentration of ^{226}Ra. The chemical toxicity was evaluated in terms of lifetime average daily dose (LADD) and hazard quotient (HQ). Hazard quotient is the risk of the chemical toxicant being characterized and it can be estimated as the ratio of the LADD of the chemical to the reference dose (RfD) [36]. The RfD is an estimate of the daily exposure to human population including vulnerable subgroups that is likely to be devoid of significant risk of adverse effects during a lifetime [37].

Life-time cancer risk

The lifetime cancer risk (LCR) associated with the intake of ^{226}Ra radionuclide was estimated using Eq. 10, obtained from the National Research Council [38].

$$LCR = MCL \times RC \times TWI \tag{10}$$

where MCL is the maximum contaminant level (Bq $\mathrm{L^{-1}}$), RC is the mortality risk coefficient (7.17×10^{-9}) and TWI is the total water intake (2L $\mathrm{d^{-1}} \times 365.4$ d $\mathrm{y^{-1}} \times$ average life expectancy). The life expectancy and the annual recommended adult water consumption rate in Ghana are 63.4 years and 730 L, respectively [39].

Chemical risk assessment

The chemical risk assessment was done to assess the carcinogenic risk related to the chemical toxicity of ^{226}Ra in the groundwater samples. This was evaluated using the life-time average daily dose (LADD) of radium through water intake and compared with the reference dose (RFD) or tolerable daily intake of 1.12 μg $\mathrm{kg^{-1}\ day^{-1}}$ [36]. The LADD was estimated using Eq. (11) and the average intake of water was estimated as 2 L $\mathrm{d^{-1}}$.

$$\text{Ingestion LADD of drinking water} = \frac{EPC \times IR \times EF \times ED}{AT \times BW} \tag{11}$$

where LADD is lifetime average daily dose (μg $\mathrm{kg^{-1}\ day^{-1}}$); EPC is the exposure point concentration (μg $\mathrm{L^{-1}}$); IR is the ingestion rate of water (L $\mathrm{day^{-1}}$); EF is the exposure frequency (days $\mathrm{year^{-1}}$); ED is the total exposure duration (years); AT is the average time (days); and BW is the body weight (kg). Thus, the hazard quotient was calculated using Eq. 12 below [37].

$$\text{Hazard quotient} = \frac{LADD}{RFD} \tag{12}$$

Data analysis

The data obtained from the sample analysis including the radio-chemical risk parameter estimates were further

evaluated using statistical tools notably multivariate and multicriteria decision-making techniques. These techniques encapsulate principal component analysis (PCA), preference ranking organisation method for enrichment evaluation (PROMETHEE) and geometrical analysis for interactive aid (GAIA). StatistiXL Version 1.8 and Visual PROMETHEE Academic Edition Version 1.4.0.0 Software were employed to undertake the statistical analysis in this study as specified in Doyi et al. [7], Gbeddy et al. [40]. The application of these statistical approaches ensured that latent information associated with the study data were critically explored in terms of pollutants interaction between sample media, pattern recognition and ranking of polluted sites.

Results and discussion

Univariate evaluation of radionuclide activity concentration

Radionuclide activity concentration in soil

The mean activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K radionuclides in the major investigatory boreholes are 159.26 Bq kg^{-1}, 92.70 Bq kg^{-1}, 1025 Bq kg^{-1}, and 172.75 Bq kg^{-1}, 91.15 Bq kg^{-1}, 1025 Bq kg^{-1} for BH1 and BH2, respectively. The mean activity concentrations of ^{232}Th, ^{226}Ra and ^{40}K in BF, a major monitoring borehole are 256.66 Bq kg^{-1}, 174.54 Bq kg^{-1}, and 1624 Bq kg^{-1}, respectively. All recorded mean values in the study were higher than the recommended world average values of 35, 45 and 420 BqKg^{-1} for ^{226}Ra, ^{232}Th and ^{40}K, respectively [3]. In this regard, it is prudent to minimize human exposure to the soil in the study area.

The mean radionuclide activity concentration in soil follows the ascending order of ^{226}Ra < ^{232}Th < ^{40}K as observed in Fig. S2. The higher activity concentration of ^{232}Th than ^{226}Ra may be due to high geological retention, and low solubility and migration of thorium into groundwater. The lowest activity concentration of radionuclides was detected at the deepest depth of 145 m in one of the key investigatory boreholes, BH1. The geological formation at the 145 m depth may be a key factor in the observed low radionuclides levels. However, the BF (BNARI Farm) borehole exhibited marginal increase in radionuclides activity concentration, with increasing depth (see Fig. S2). It must be noted that BF is a shallow depth borehole compared to BH1 and BH2. The abundance of ^{40}K constituting about 86% of the total radionuclide (^{226}Ra + ^{232}Th + ^{40}K) concentration reflects the abundance of potassium silicate minerals, principally alkali feldspars and micas in the geological formation of the study area. Moreover, the intense agricultural activities in the study area with the attendant use of potassium containing fertilizers may also contribute to the high levels of ^{40}K radionuclide.

Radionuclide activity concentration in groundwater

The activity concentration of ^{232}Th, ^{226}Ra, and ^{40}K radionuclides ranged from 0.52 to 8.25, 0.33–3.28, and below detection limit (BDL)—6.53 Bq L^{-1}, respectively (see Table S4). The mean values for ^{232}Th and ^{226}Ra are 1.67 ± 0.42 and 1.45 ± 0.42 Bq L^{-1}, respectively. From Fig. S3, ^{40}K has the highest activity concentration across most sampling sites which is akin to the radionuclide's distribution in soil matrix. However, the groundwater from B-MG borehole has an elevated activity concentration of ^{232}Th which may be due to the differences in geological formation underlying the study sites. All the study sites have ^{40}K activity concentration below the World Health Organization (WHO) recommended permissible level of 10 Bq L^{-1}. However, most of the boreholes, in exception of those situated at the Graduate School of Nuclear and Allied Sciences (SNAS) and GAEC Guest House (GGH) have ^{232}Th and ^{226}Ra activity concentrations above WHO's permissible level of 1 Bq L^{-1}. The high concentrations of ^{232}Th and ^{226}Ra radionuclides in the groundwater can be ascribed to their high concentrations in the bedrock [4]. The relatively high activity concentration of ^{226}Ra compared to ^{232}Th in the groundwater can also be attributed to their higher solubility in groundwater [41].

The results from the univariate evaluation of the radionuclide activity concentrations indicate the preponderance of ^{40}K in both soil and groundwater samples. In order to determine the existence of radionuclides interaction between soil and groundwater matrices, the activity data in Tables S2 and S4 was further evaluated using multicriteria decision-making approach.

Geological interaction of soil and groundwater radionuclides

The activity concentration data matrix consisting of 14 actions × 3 criteria was subjected to preference ranking organisation method for enrichment evaluation (PROMETHEE) and geometrical analysis for interactive aid (GAIA). A pairwise comparison in all plausible combinations was done for the 42 entries in the data matrix by subtraction thus resulting in a difference for each comparison [42]. All the criteria were assigned equal weights and V− shape preference function was selected. Negative outranking flow (*Phi*-) and positive outranking flow (*Phi*+) were estimated in order to guide the preference selection process. *Phi*- specifies how each action is outranked by all others whilst *Phi*+ describes how each action outranks all others, and the difference between *Phi*+ and *Phi*- is referred to as the net outranking flow, *Phi* (that is *Phi* = (*Phi*+) − (*Phi*−))

[42]. A complete ranking (PROMETHEE−II) was undertaken using the *Phi* values with preference for minimal radionuclide activity concentration. This approach enabled the complete ranking of samples in terms of radionuclide concentration (from least to maximum pollution) as shown in Table 1, and also helps identify patterns in the data for radionuclides interaction assessment as depicted by the GAIA biplot, Fig. 1.

Table 1 Complete ranking (PROMETHEE-II) of radionuclides activity concentrations in soil and groundwater samples

Rank	Sample ID	Phi	Phi+	Phi−
1	BH2g	0.7977	0.7977	0.0000
2	BH1g	0.7964	0.7965	0.0002
3	BH1,145	0.4382	0.5116	0.0734
4	BH2,75	0.2104	0.3474	0.1370
5	BH2,148	0.1382	0.2898	0.1516
6	BH1,5	0.0976	0.2674	0.1698
7	BH1,75	0.0865	0.2617	0.1752
8	BH2,120	− 0.1433	0.1407	0.2840
9	BH1,50	− 0.1851	0.1399	0.3250
10	BH2,100	− 0.2342	0.1053	0.3395
11	BH2,50	− 0.4575	0.0390	0.4965
12	BH1,100	− 0.4779	0.0368	0.5147
13	BH1,120	− 0.5079	0.0233	0.5312
14	BH2,5	− 0.5590	0.0210	0.5799

Sample ID's ending with g denote groundwater whilst the numbers after the comma in the sample ID represent the depth (in metres) at which soil sample was taken. Negative outranking flow (Phi-) and positive outranking flow (Phi+) were estimated in order to guide the preference selection process. Phi- specifies how each action is outranked by all others whilst Phi+ describes how each action outranks all others, and the difference between Phi+ and Phi− is referred to as the net outranking flow, Phi (that is Phi = (Phi+)− (Phi−)) [42]. Ranking is in ascending order of radionuclide activity concentration

The biplot in Fig. 1 shows that the soil and groundwater samples segregate into two major clusters. One of the clusters consists of granulated rock samples (soil) from 5, 50, 100 and 120 m depths from boreholes BH1 and BH2 located at the left-half of the diagram. This cluster constitutes a class of samples with high radionuclide activity concentration by virtue of their opposite position to the pi-decision axis (that is the thick red line on the positive U axis of the biplot). Moreover, according to the PROMETHEE−II ranking in Table 1, this cluster of samples exhibited the highest activity concentrations since they are ranked higher in the table.

The second cluster is made up of soil samples from 5, 75, 145 and 148 m depth, and groundwater from boreholes BH1 and BH2 (see Fig. 1). This cluster represents the class of samples with the least radionuclide activity concentration by virtue of their proximity to the pi-decision axis as well as the least ranking on Table 1. The activity concentrations in the deepest depth of the two key boreholes, 145 m and 148 m are highly correlated with groundwater samples as well as having the greatest proximity to the pi-decision axis. In this regard, the deepest depth geological formations and groundwater samples have the least activity concentration of ^{232}Th, ^{226}Ra, and ^{40}K radionuclides. The lithology of the study area as indicated in Table S2 shows the existence of massively dark coloured silicified schist formations at the

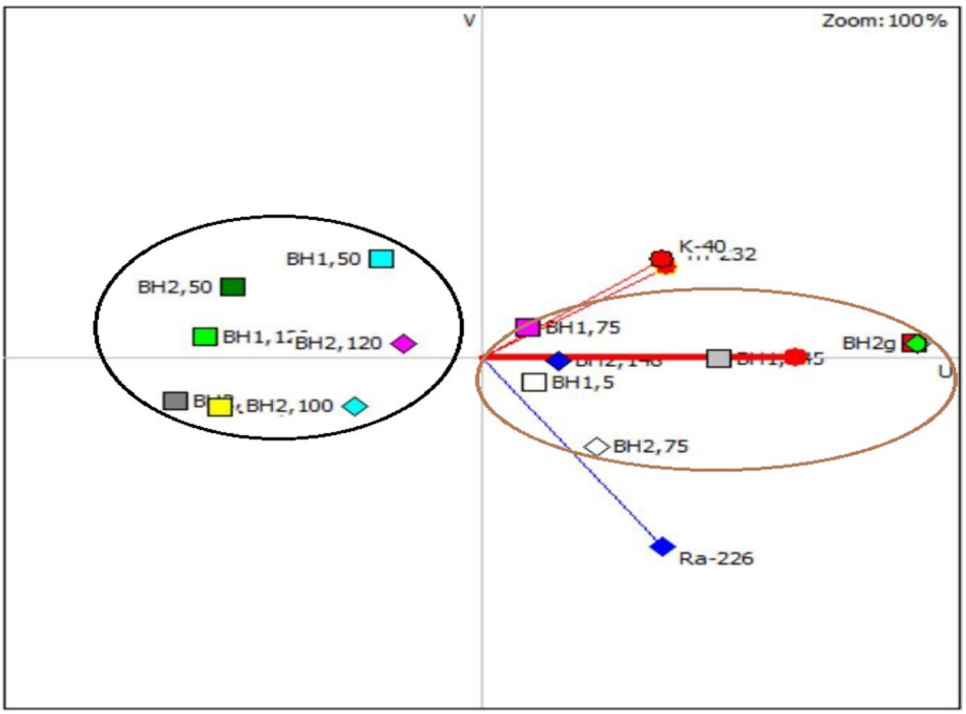

Fig. 1 GAIA biplot for radionuclides activity concentrations in soil and groundwater samples

deepest depth of the boreholes, thus indicating potential minimal availability of primordial or terrestrial radionuclides. Potential hydro-geological interaction of radionuclides exists between the rock (soil) and groundwater media at the deepest depth of the boreholes. Considering the linear pattern of distribution of the radionuclide activity concentrations in the groundwater and the deepest depth rock samples from the boreholes as shown in Fig. 1, it can be inferred that an equilibrium condition has been attained in the migration of radionuclides between the two media.

From Fig. 1, ^{232}Th and ^{40}K radionuclides are highly correlated thereby indicating similar potential terrestrial geological origin of these radionuclides. However, ^{232}Th and ^{40}K are orthogonal to ^{226}Ra, thus, demonstrating dissimilar primordial rock or soil origin of these two categories of radionuclides. The geological conditions of an area is established to be a determining factor in the natural radioactivity of the environment [24, 43]. The study area is characterized by varied mineral composition of rocks at various depths as evident in the lithological structure in Table S2, thus influencing the origins of these radionuclides. ^{232}Th and ^{40}K radionuclides are closer to the pi-decision axis than ^{226}Ra. In this context, ^{232}Th and ^{40}K exhibited more influence on the distribution of radionuclides in the study area.

Radio-chemical risk assessment of radionuclides

The activity concentrations of ^{232}Th, ^{40}K and ^{226}Ra in soil and groundwater samples were used to determine the baseline radiation doses posed by these terrestrial radionuclides to the public prior to the actual implementation of the borehole disposal system (BDS) in Ghana. Considering the intense negative perception of the populace to the nuclear industry and radiation related issues, it is fundamental to assure the public that all the necessary radiation risk assessment pre-requisites are met prior to the deployment of the new concept of BDS. Secondly, the IAEA has a vested interest in the implementation of the BDS since Ghana is one of the pioneering countries in the application of the BDS technology. In this regard, both local and international safety assessment protocols need to be met in order to satisfy all the necessary regulatory demands.

Risk assessment of radionuclides in soil

Based on existing literature, six (6) radio-chemical risk assessment parameters involving the absorbed dose rate (AD), annual effective dose equivalent (AEDE), radium equivalent activity (Ra_{eq}), activity utilization index (AUI), annual gonadal equivalent dose (AGED), and excess lifetime cancer risk (ELCR) were estimated for the radionuclides in soil. The results of the radio-chemical risk assessment are shown in Table 2. To eliminate highly correlated and redundant risk parameters thereby prioritizing risk assessment in case of an emergency, the 17 observations × 6 risk parameters data matrix in Table 2 was subjected to principal component analysis (PCA). The data was normalized using Kaiser–Meyer–Olkin normalization due to the differences in the units of the parameters.

The results of the PCA in Table 3a showed that only one principal component (PC) with Eigenvalue of 5.98 accounting for 99.7% variability in the data was found to be significant, based on an Eigenvalue > 1 [44]. As a result, the biplot was not considered, however, the correlation matrix and the factor loadings for the PCs are indicated in Table 3b and c, respectively. Although, all the parameters were estimated using different formula, the PCA results show significant correlation (correlation coefficients > 0.99 at 0.95 significant level) among all the variables. Notably, the correlation coefficient between AGED and AD, and AUI and Ra_{eq} are equal to 1. Thus, it may not be necessary to estimate all six risk parameters during radio-chemical risk assessments. The annual gonadal equivalent dose (AGED), however, contributes the highest loading to PC1 (see Table 3c). In this regard, AGED is considered as the most representative radio-chemical risk parameter for terrestrial radionuclides in soil, hence it merits further discussion.

From Table 2, the estimated AGED values for the soil samples ranged from 618.78 to 2,889.95 μSv y^{-1}, with an average of 1,561.69 μSv y^{-1}. The reported world average value for AGED is 300 μSv y^{-1} for soil [3]. All the estimated baseline AGED values in this study exceeded the allowable limit, thus, signifying the existence of significant radio-chemical risk in the study area prior to the implementation of the BDS concept. Therefore, the probability of human exposure to terrestrial radionuclides, ^{232}Th, ^{40}K and ^{226}Ra in the soil has to be drastically reduced to forestall any potential stochastic radio-chemical effects such as gene mutation and cancer.

Risk assessment of radionuclides in groundwater

Four (4) radio-chemical risk parameters entailing age-dependent annual effective ingestion dose (AAEID), life-time cancer risk (LCR), life-time average daily dose (LADD) and hazard quotient (HQ) were evaluated based on prevailing literature. The AAEID was considered for infants, children, and adults for each radionuclide. Data matrix made up of 14 observations × 12 variables (see Table S5) was analyzed using principal component analysis (PCA). The data was pre-processed similar to the soil data as a result of varied units of variables.

From Table 4a, three (3) principal components have been significantly extracted based on Eigenvalues > 1 and they account for approximately 99.8% of the variance in the data. The factor loadings for the variables are indicated in

Table 2 Radio-chemical risk assessment of radionuclides in soil

Sample ID	Radio-chemical risk parameters					
	AD (nGy/h)	AEDE (mSv/y	Ra_{eq} (Bq/kg)	AUI	AGED (μSv/y)	ELCR (mSv/y)
BH1,5m	150.62	0.18	331.91	2.36	1057.65	0.0006
BH1,50m	189.64	0.23	416.31	3.09	1322.17	0.0008
BH1,75m	145.17	0.18	318.05	2.28	1017.19	0.0006
BH1,100m	263.67	0.32	582.29	4.17	1850.72	0.0011
BH1,120m	253.47	0.31	553.87	3.91	1778.56	0.0011
BH1,145m	88.14	0.11	194.01	1.38	618.78	0.0004
BH2,5m	286.75	0.35	636.19	4.64	2008.93	0.0012
BH2,50m	242.23	0.3	530.1	3.84	1694.12	0.001
BH2,75m	125.57	0.15	271.9	1.73	892.2	0.0005
BH2,100m	207.09	0.25	452.91	3.12	1458.51	0.0009
BH2,120m	183.11	0.22	399.67	2.8	1285.95	0.0008
BH2,148m	135.46	0.17	295.05	2.04	953.06	0.0006
BF,9m	182.37	0.22	405.3	3.03	1272.91	0.0008
BF,27m	264.54	0.32	575.99	3.98	1860.8	0.0011
BF,45m	295.25	0.36	645.81	4.67	2065.17	0.0013
BF,60m	360.25	0.44	791.35	5.72	2522.13	0.0015
BF,72m	414.56	0.51	914.72	6.84	2889.95	0.0018

Where the numbers after the comma in the sample ID represent the depth (in metres) at which soil sample was taken. AD, AEDE, Ra_{eq}, AUI, AGED, and ELCR refer to absorbed dose rate, annual effective dose equivalent, radium equivalent activity, activity utilization index, annual gonadal equivalent dose, and excess lifetime cancer risk, respectively

Table 3 PCA Eigenvalues (a), Spearman correlation matrix (b) and factor loadings (c) for radio-chemical risk parameters in soil

(a)

	PC1	PC2	PC3	PC4
Variability (%)	99.655	0.166	0.163	0.015
Cumulative %	99.655	99.821	99.985	100.000

(b)

Variables	AD	AEDE	Ra_{eq}	AUI	AGED	ELCR
AD	**1**					
AEDE	**0.998**	**1**				
Ra_{eq}	**0.995**	**0.998**	**1**			
AUI	**0.995**	**0.998**	**1.000**	**1**		
AGED	**1.000**	**0.998**	**0.995**	**0.995**	**1**	
ELCR	**0.993**	**0.994**	**0.993**	**0.993**	**0.993**	**1**

(c)

	PC1	PC2	PC3	PC4
AEDE	1.000	−0.010	0.000	0.027
Ra_{eq}	0.999	−0.020	0.050	−0.006
AUI	0.999	−0.020	0.050	−0.006
AGED	0.999	−0.020	−0.050	−0.006
ELCR	0.996	0.091	0.000	−0.002

Values in bold are different from 0 with a significance level alpha = 0.95

Table 4b whilst the biplot is shown in Fig. 2, and the correlation matrix is stipulated in Table S6 of the Supplementary Information.

From Fig. 2, the risk parameters for radionuclides in groundwater segregated into three (3) groups. The age-dependent annual effective ingestion dose (AAEID) due to ^{40}K exposure in infants (K-40(i)), children (K-40(c)), and adults (K-40(a)) form one cluster as evident in the high correlation between these parameters in Table S6. However, K-40(c) can be considered as the representative risk parameter in this cluster due to its relatively higher contribution to PC1 (see Table 4b). In this regard, there is no need to estimate each risk parameter especially during time constraint situations.

The AAEID for ^{232}Th radionuclide exposure in infant (Th-232(i)), children (Th-232(c)), and adults (Th-232(a)) also constitute another cluster as apparent in the significant correlation among these parameters in Table S6. Any variable in this cluster can potentially represent the risk posed

Table 4 PCA Eigenvalues (a) and factor loadings (b) for radio-chemical risk parameters in groundwater

	PC1	PC2	PC3	PC4	PC5
(a)					
Eigenvalue	7.635	3.200	1.143	0.019	0.003
Variability (%)	63.621	26.668	9.525	0.160	0.025
Cumulative %	63.621	90.289	99.815	99.975	100.000
	PC1	PC2	PC3	PC4	PC5
(b)					
Th-232(i)	0.817	0.285	0.502	0.000	0.000
Ra-226(i)	0.969	−0.133	−0.209	0.001	0.000
K-40(i)	0.027	0.976	−0.202	−0.069	−0.036
Th-232(c)	0.817	0.285	0.502	0.000	0.000
Ra-226 (c)	0.969	−0.133	−0.209	0.001	0.000
K-40(c)	0.036	0.974	−0.216	−0.042	0.042
Th-232(a)	0.817	0.285	0.502	0.000	0.000
Ra-226(a)	0.969	−0.133	−0.209	0.001	0.000
K-40(a)	0.014	0.974	−0.194	0.112	−0.006
LCR	0.969	−0.133	−0.209	0.001	0.000
LADD	0.969	−0.133	−0.209	0.001	0.000
HQ	0.969	−0.133	−0.209	0.001	0.000

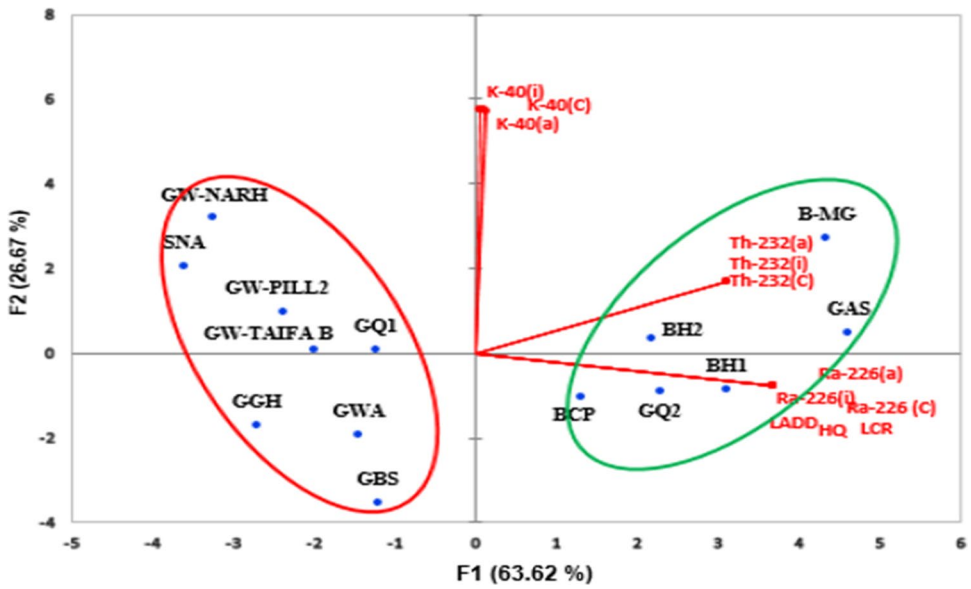

Fig. 2 PCA for groundwater radio-chemical risk parameters

by this radionuclide in groundwater due to the identical factor loadings in PC1, PC2 and PC3 (see Table 4b).

The third cluster is made up of the AAEID for ^{226}Ra radionuclide exposures in infants (Ra-226(i)), children (Ra-226(c)), adults (Ra-226(a)), life-time cancer risk (LCR), life-time average daily dose (LADD), and hazard quotient (HQ). These parameters are highly correlated as shown in Table S6. Any of the variables in this cluster can be considered as a representative parameter due to their identical factor loadings in PC1, PC2 and PC3 as shown in Table 4b. LADD, LCR and HQ are related to radium (Ra) ingested health hazards, thus, the observed significant correlation among these parameters.

Furthermore, from Fig. 2 the various groundwater samples partitioned into two classes. One class is made up of BH1, BH2, BCP, GQ2, B-MG and GAS in the right side green circle of the biplot. The estimated radio-chemical risk variables are highly correlated with these samples, thus indicating significant influence of these risks in the groundwater at these sites. From the map of the study area as shown in Fig. S1, these boreholes are located within the GAEC site which hosts the Ghana nuclear research reactor, gamma irradiation facility and the centralized radioactive waste storage facilities. This observed correlation may be attributed to the conduct of these nuclear related activities over the years which could emanate in elevated levels of radiation beyond the background concentration. This inference is based on the fact that the groundwater samples within the left red circle in the biplot of Fig. 2 constituting the second class of samples are not correlated with the radio-chemical parameters. This second class of samples are located far away from the GAEC site as indicated in Fig. S1. In this regard, the conduct of nuclear related activities may not have any direct effect on the groundwater radioactivity concentrations coupled with potential differences in the geologic formations of the areas associated with both classes.

Most of the estimated AAEID derived from the ingestion of ^{226}Ra and ^{232}Th are comparatively higher than UNSCEAR's reference individual dose range of 0.2–1 mSv y^{-1} for radionuclides of natural origin [3, 45]. This indicates that the groundwater may not be suitable for long-term consumption since the ingestion of radionuclides such as ^{226}Ra in the water could lead to deleterious health effects [34]. In this regard, safety precautions such as limiting access to the groundwater resources in the study area must be applied coupled with intense public education. Although ^{226}Ra is considered a primordial radionuclide, anthropogenic activities such as the emplacement of conditioned radioactive waste into the host rock could lead to elevated activity concentrations and radio-chemical risk of Ra if the disposal system is not adequately engineered.

Conclusion

The activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K in soil and groundwater from boreholes located at potential geological disposal site within the Ghana Atomic Energy Commission and its catchment were measured using gamma spectrometry equipped with high resolution hyper pure germanium (HPGe) detector with a relative efficiency of 30% relative to a 3″×3″ NaI (Tl) scintillator. The results obtained were used to evaluate the geological interaction of radionuclides and radio-chemical health risk posed to humans across the study area with the aid of multivariate and multicriteria decision-making analytical tools. Most of the boreholes have ^{226}Ra and ^{232}Th activity concentrations above WHO's permissible level of 1 Bq L^{-1}. The results from principal component analysis (PCA), preference ranking organisation method for enrichment evaluation (PROMETHEE) and geometrical analysis for interactive aid (GAIA) indicate that groundwater and soil separate into two major clusters based on the depth of samples. The PROMETHEE and GAIA results indicate that the activity concentrations at soil depth of 145 m and 148 m are highly correlated with groundwater samples, thus, indicating similar origin and potential geological interaction of radionuclides. From the PCA results, all the six radio-chemical risk parameters assessed in soil were highly correlated, hence lessening the need to individually determine these parameters and reducing cost and time of risk assessment. On the other hand, the twelve risk parameters estimated for groundwater segregated into three clusters and the clusters were strongly influenced distinctively by each of ^{40}K, ^{226}Ra, and ^{232}Th radionuclides. The conduct of nuclear related activities over the years may influence the radio-chemical risk of groundwater at the GAEC site. This study has, therefore, established the terrestrial radioactivity concentration and associated radio-chemical risks using relevant statistical approaches to facilitate informed decision making during the implementation of the borehole disposal system both locally and internationally. This research calls for persistent investigation of groundwater samples within areas associated with nuclear activities in order to protect the environment and human health thereby ensuring that unnecessary burden is not placed on future generations.

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Declarations

Conflict of interest The authors declare that they have no conflict of interest.

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