



Natural radioactivity in some geological formation in the Accra Plains

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ABSTRACT

The activity concentrations of natural occurring radionuclides, uranium (²³⁸U), thorium (²³²Th), radium (²²⁶Ra) and potassium (⁴⁰K) were measured in geological samples collected from the Accra Plains using a high resolution hyper pure germanium (HPGe) detector. Their radiological parameters were also calculated. ²³⁸U concentration was found to vary from below detection limit to 363.5 Bq/kg. Activity concentration of ²³²Th varied from below detection limit to 370.41 Bq/kg. The activity concentration of ²²⁶Ra ranged from 4.04 Bq/kg to 295.01 Bq/kg, whereas, the ⁴⁰K activity concentration varied from 145 Bq/kg to 2274.3 Bq/kg. The absorbed dose rates varied from 18.27 to 479.26 nGy/h. The annual external effective dose rates ranged from 0.02 to 0.51 mSv/y. The radium equivalent activity (Raeq) varied from 29.15 to 986.42 Bq/kg. Value of external hazard index (Hex) varied from 0.11 to 2.66. Good correlations existed between ²³²Th and ²²⁶Ra activity concentrations and the total activity concentration. The Th/U ratios obtained indicated metasomatic activity of the analyzed radionuclides. The gamma ray radiation hazards due to the radionuclides increased with depth.

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Introduction

There is international consensus, within the nuclear industry and among governments, that emplacement of radioactive waste in geological repository is the best long term waste management option that is currently available (Sumerling and Smith, 1998). Geological repository is a facility for radioactive waste disposal located underground (usually from a few tens of meters to several hundred meters below the surface) in a stable host rock to provide long term isolation of radionuclides from the accessible environment (IAEA, 2003). Disposal of radioactive wastes in geological repository will add a concentrated source of radioactivity to the natural radioactivity dispersed in the geological formation.

Natural sources of radiation are the concentration of natural occurring radionuclides that represent ambient conditions present in the environment that are in no way influenced by human activity (Egidi, 1998). According to the United Nations Scientific Committee on Effects of Atomic Radiation (UNSCEAR) 2000 report, natural sources of radiation are the largest contributors (87%) to the external dose of the world population. The worldwide average annual effective dose from natural background radiation is 2.4 mSv (UNSCEAR, 2000). Naturally occurring uranium (²³⁸U) and thorium (²³²Th) are of particular interest in radioactive waste disposal since they represent useful analogues of the actinides (Smellie et al. 1986) and make the biggest contribution to the total background dose. In addition to being the main source of continuous radiation exposure to human, natural sources of radiation is the basic indicator of radiological contamination in the environment. Their high geochemical mobility allows them to be easily mobilized into the environment (Faanhof, 1999).

Terrestrial radioactivity depends mainly on local geological and geographical conditions, and it is especially related to the rock types (Florou and Kritidis, 1992). Their distributions

depend on the distribution of rocks from which they originate, lithologic character and the processes which result in their removal from the soil and migration (Anjos et al., 2005; Arafa, 2004). Higher radiation levels are associated with igneous rocks, such as granite, and lower levels with sedimentary rocks. According to Roger and Adams (1969) uranium and thorium are generally enriched in the youngest, most felsic and potassic members of comagmatic suites of igneous rocks. There are exceptions, however, as some shales and phosphate rocks have relatively high content of radionuclides (UNSCEAR Report, 1993).

Precambrian crystalline bedrock is one of the geological formations considered as a potential host rock for safe disposal of radioactive wastes. There is a variety of crystalline (i.e. igneous or metamorphic) rocks. They include granites, granodiorites, gneisses, gabbro, even basalt.

Knowledge of the dose received from natural radioactivity can serve as a standard reference to evaluate the incidence of radiation from man-made sources. This study was carried out to establish the abundance and distribution of U, Th and K in the geological formation of the Accra Plains and also to determine the radiation doses from the activity concentrations

Geochemistry of Uranium, Thorium and Potassium

Naturally occurring uranium is widespread in the environment and consists of three isotopes, all of which are radioactive: ²³⁸U (99.2739%, half life 4.5 x 10⁹ years), ²³⁵U (0.7025%, half life 7.3 x 10⁸ years) and ²³⁴U (0.0057%, half life 2.48 x 10⁵ years). The ratio of ²³⁵U to ²³⁸U is less than 1%, hence the contribution of ²³⁵U to the environmental dose is very small (El-Dine et al., 2001). ²³⁴U is a decay product of the ²³⁸U series. Most crustal rocks contain uranium averaging about 2.7 ppm or 3.7 Bq/kg (UNSCEAR 2000). The decay product of ²³⁸U, ²¹⁴Pb is measured by gamma-ray spectrometry because it is directly

related to U radioactivity. The dominant uranium valence states that are stable in geologic environments are uranous (U^{4+}) and uranyl (U^{6+}) states with uranyl being more soluble than the uranous (NRC, 1999).

Naturally occurring thorium as found in nature is 100% ^{232}Th by weight, although other isotopes such as ^{230}Th , a part of the uranium decay series, are also found in nature. Its decay product ^{208}Tl is measured by gamma-ray spectrometry because it is directly related to thorium radioactivity. Thorium is also widely found in crustal rocks, being more abundant in acidic materials than alkaline ones. By weight the average thorium content of the continental upper crust and soils is about 9-10 ppm, about four-fold greater than that of uranium (Kathren, 1998). However, thorium has a much lower specific activity than uranium and the radioactivity concentration of the two elements is about the same.

Both thorium and uranium decay series include isotopes of radium (Ra). The isotope ^{226}Ra is particularly important. It decays by alpha emission to ^{222}Rn (radon). ^{222}Rn has a half-life of 3.82 days and is chemical inert gas. The principal hazards posed by U and Th are their chemical toxicity and the radiological hazards posed by some of their decay products, especially those that are alpha particle emitters (^{226}Ra and ^{222}Rn) which are of concern if they are inhaled or ingested.

Potassium has 24 known isotopes three of which occur naturally: ^{39}K (93.3%), ^{40}K (0.0117%) and ^{41}K (6.7%). Potassium-40 (^{40}K) is radioactive and has a half-life of 1.28×10^9 yr and gives specific activity of 31.4Bq/g of natural potassium (Kathren, 1998). ^{40}K decays to stable ^{40}Ar (11.2%) by electron capture and by positron emission resulting in the release of gamma photons with maximum energy of 1.46 MeV. ^{40}K also undergoes decay to stable ^{40}Ca (89%), emitting a 1.314 Mev beta particle in the process. For crustal rock, the mean ^{40}K activity is 0.62Bq/g with basalt averaging about half the concentration. Potassium (K) is a major constituent of many rock forming minerals (K-feldspars - orthoclase, microcline), and is enriched in acidic igneous rocks relative to basic ones. The K_2O content of K-feldspars varies from 11 to 15% while in biotite and muscovite it varies from 8 to 10% and from 10 to 11%, respectively.

Both uranium and thorium exist in the +4 oxidation states in most geologic environment. However uranium unlike thorium has the possibility of either a pentavalent or a hexavalent state. The pentavalent state of uranium, like the trivalent state of thorium or uranium is not expected under geologic conditions (Seaborg and Katz, 1954). Under reducing conditions, uranium and thorium are stable and difficult to migrate. However, their high valence compounds and complexes have low melting points, small densities, and are water-soluble. Therefore, under oxidative conditions, they are prone to migration in the environment (Xuezhao and Ali 1998).

Study Area

The Accra Plains is located in the south eastern part of Ghana between latitude $5^{\circ} 30'$ and $6^{\circ} 15'$ North of the equator and longitude $0^{\circ} 20'$ west and $0^{\circ} 40'$ east of the Greenwich meridian. Geologically, greater part of the Plains is underlain by very old crystalline metamorphic rocks (Dahomeyan gneiss) which belong to the Pan-African terrain called Dahomeyides (Fig.1)(Attoh, 1998).

The Dahomeyan gneisses are divided into felsic and mafic gneisses. The mafic gneisses are composed of thick series of mafic rocks rich in ferro-magnesian minerals consisting of rock

types such as garnet hornblende gneiss (dominant), garnet-hornblende-pyroxene gneiss, garnet-pyroxene gneiss with a few minor layers of hornblende and biotite schist and gneiss at the base (Kesse 1985).

The felsic gneiss and composite group of rocks are composed of quartz, feldspar, epidote and mica, although some amphibolites occur locally. Hornblende and garnet, in addition to quartz, feldspar, epidote and mica are the principal minerals of the rocks of the composite groups.

Other geological units in the Plains include the Togo series, the Accraian series, the Tertiary and Recent sediments. The Togo series consist mainly of quartzites, phyllites, sandstones, schists and silicified limestones. They occur as outliers in the Plains. The Accraian series consisting of sandstones, grits and shales are found near Accra. The Tertiary and recent sediment occur in the south eastern part of the Plain near the estuary of the Volta River.

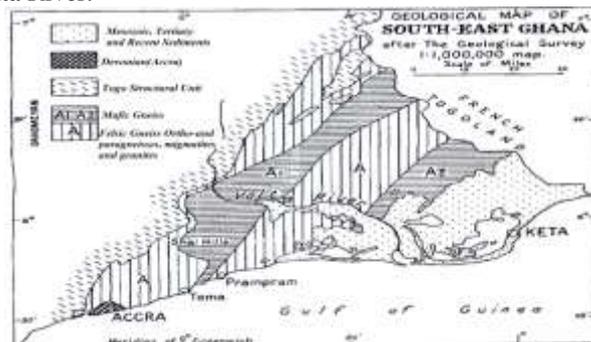


Fig1. Geological map of the Accra Plain
Sampling and sample preparation

A total of thirty eight (38) geological samples were collected from six (6) different locations in the Accra Plains. Twenty two (22) of the samples (BF) were collected from a borehole drilled at the Ghana Atomic Energy Commission premises at Kwabenya. The samples were collected at 3m interval from a depth of 9m to 72m. Two (2) samples (ASH) were collected from two different locations at Ashiaman at depths of about 5m. Three (3) shale (S) soil samples were also collected from a borehole drilled near the Afua Sunderland Children's Park in Accra at depths ranging from 5 to 20 m. In Tema, six (6) samples (RD) were collected from four (4) boreholes drilled (4 -15.5m deep) at different points at the Ghacem Factory premises. Four (4) samples (outcrops) were also collected from different points around the foot of the Krobo Mountain (KD). A clay sample (CK) was also collected from Okwenya.

All the samples were oven dried at about $110^{\circ}C$ (IAEA, 1989). The samples were then crushed, grinded, homogenized and sieved to $250\mu m$ grain size. 70g of each sample was transferred into an airtight cylindrical plastic container with a volume of $250cm^3$. The samples were sealed and stored for more than 30days before analysis. This allowed the in-growth of uranium and thorium decay products and prevented the escape of radiogenic gases ^{222}Rn and ^{220}Rn as well as allowed secular equilibrium to be reached between ^{238}U , ^{232}Th and their daughter products.

Experimental Set-up

The concentrations of the radionuclides in the sample were determined by employing a high resolution hyper pure germanium (HPGe) detector with a relative efficiency of 30% relative to a $3'' \times 3''$ NaI (TI) scintillator. The energy resolution

(FWHM) of the detector was 2keV at 1.332MeV of a ^{60}Co source. The detector was placed in a lead shield to reduce the background radiation originating from the building materials and cosmic rays surroundings. Advanced multi-channel analyzer (MCA) emulation software (MAESTRO) was used for data acquisition, storage, display and online analysis of the acquired gamma-spectra.

Each sample was put into the shielded HPGe detector and measured for an accumulating time of about 16 h. Prior to measurement of the samples, the environmental gamma background in the laboratory was determined with an empty cylindrical container of the same geometry as the sample containers, under identical measurement conditions. The measured background activity was subtracted from the measured activity of each sample.

The gamma ray transitions of energies 186.3 keV (^{226}Ra), 351.9keV (^{214}Pb), 609.3keV(^{214}Bi), 768.4 keV and (^{214}Bi) were used to determine the ^{238}U series concentrations. The gamma-ray lines at 911.0 keV(^{228}Ac), and 583.3 keV(^{208}Tl) were used to determine the concentration of the ^{232}Th series. ^{40}K concentration was determined using the peak area at 1460keV. The 661.66keV gamma transition was used for the determination ^{137}Cs concentration.

Calculation of activity concentration

Calculation of counts rates for each detected photopeak and radiological concentrations (activity per mass unit) of detected radionuclides depend on the establishment of secular equilibrium in the samples. Since secular equilibrium was reached between ^{232}Th and ^{238}U and their decay products, the ^{232}Th concentration was determined from the average concentrations of ^{208}Tl and ^{228}Ac in the samples. The ^{238}U concentrations were determined from the average concentrations of the ^{226}Ra , ^{214}Pb and ^{214}Bi decay products. A true measurement of ^{226}Ra , ^{40}K and ^{137}Cs concentrations were made. The specific activity in Bq/kg was calculated using the equation

$$A_{Ei} = \frac{N_{Ei}}{\epsilon_E \times t \times \gamma_d \times M_s} \quad (1)$$

where N_{Ei} is the net peak area of a peak at energy E , ϵ_E the detection efficiency at energy E , t the counting live-time, γ_d the gamma ray yield per disintegration of the specific nuclide for a transition at energy E and M_s the mass of the measured sample in kilogram (kg).

Radiological concentrations of ^{232}Th , ^{238}U , ^{226}Ra and ^{40}K were converted into total elemental concentrations of thorium, uranium, radium and potassium respectively according to the expression

$$F_E = \frac{M_E C}{\lambda_E N_A f_{A,E}} \frac{1}{n} \sum_{i=1}^n A_i \quad (2)$$

where F_E is the fraction of element E in the sample, M_E is the atomic mass (kg mol^{-1}), λ_E is the decay constant (s^{-1}) of the parent isotope, N_A is Avogadro's number (6.023×10^{23} atoms mol^{-1}), $f_{A,E}$ is the fractional atomic abundance of ^{232}Th , ^{238}U , ^{226}Ra or ^{40}K in nature, C is a constant that converts the ratio of the element's mass to soil mass into a percentage or ppm and A_i is the radiological concentration of ^{40}K ($n=1$) or that of selected daughter radionuclides in the decay series of ^{232}Th and ^{238}U ($n=2$).

The total uncertainty (σ_{tot}) of the calculated activity values is composed of the counting statistical (σ_{st}) and weighted systematic errors ($\sigma_{sys,i}$) calculated by the following formula

$$\sigma_{tot} = \sqrt{\sigma_{st}^2 + \frac{1}{3} \sum_i \sigma_{sys,i}^2} \quad (3)$$

The systematic uncertainties considered include, the uncertainty of the source activity (3%), the uncertainty in the efficiency fitting function (1-10%) and uncertainties in the nuclide master library used. The total uncertainty in the calculated activity concentration for the rock and soil samples was below 5%.

Results and Discussion

The descriptive statistics of the activity concentrations of the samples are presented in table 1 whilst the relative abundance of the samples is presented in table 2.

The BF, ASH and S had higher ^{238}U concentration than the worldwide mean ^{238}U concentration of 35Bq/kg for rock/soil (UNSCEAR 2000). The worldwide mean ^{232}Th concentration for rock/soil is 30Bq/kg indicating that the activity concentrations obtained for the BF,ASH,S and some of the RD samples had higher ^{232}Th concentrations. The ^{232}Th activity concentration in the BF samples increased with depth especially from 33m to 72m (fig.2).

The worldwide mean ^{40}K concentration reported by UNSCEAR (2000) is 400Bq/kg. Potassium concentrations in a wide variety of rock types were estimated to range from approximately 0.1 to 3.5%. The rock types investigated in this study had potassium values higher than this value except for the KD samples which had values ranging from 0.47 to 1.09%. The high ^{40}K concentration may be due to high potassium feldspar mineral contents in the samples.

^{137}Cs is a man-made high specific-activity long-lived radionuclide. It does not exist naturally in soil, sand and their source rocks but as product of radioactivity fallout. It is strongly absorbed to small soil and clay particles. The standard concentration range for ^{137}Cs ranges from 10 to 1000Bq/kg (El-Dine et al 2004). Minimum concentrations of ^{137}Cs (0.3 – 3.12 Bq/kg) were found in the samples from Ashiaman and the clay (montmorillonite) sample from Okwenya as well as in one of the KD samples.

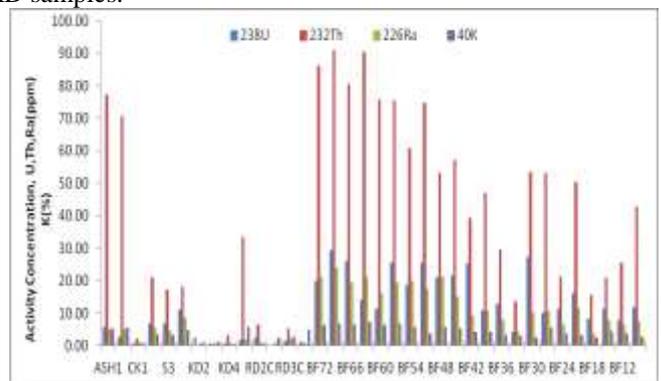


Fig.2. Activity concentration of ^{238}U , ^{232}Th , ^{226}Ra and ^{40}K in the samples

Distribution of Thorium, Uranium, and Potassium

The relationship between the thorium (Th), uranium (U), radium (Ra) and potassium (K) concentrations and the total activity concentration in the samples are diagrammatically shown in fig. 3(a-d). Good correlations exist between ^{232}Th and ^{226}Ra activity concentrations and the total activity concentration for all the samples with correlation coefficients of 0.911 and 0.912 respectively. The BF samples had correlation coefficients (R^2) of 0.932 for ^{232}Th and 0.95 for ^{226}Ra . The ^{238}U and ^{40}K activity concentrations had a fairly good correlation with the

total activity concentration with correlation coefficients of 0.712 and 0.716 respectively. This implies that ²³²Th and ²²⁶Ra contributed significantly to the total radioactivity of the samples.

The original concentrations of uranium, thorium and potassium in rocks may vary because of alteration or metamorphic processes (Verdoya et al., 2001). The Th/U, K/U, K/Th, K/Ra and Th/Ra ratios which may indicate whether relative depletion or enrichment of radionuclides had occurred are shown in fig 4(a-d). The relations between Th and U, K and U, K and Th, K and Ra, and Th and Ra have the following correlation coefficients of 0.52, 0.29, 0.63, 0.55 and 0.74 respectively. The average Th/U ratio was 4.25. The theoretically expected Th/U ratio for a normal continental crust is 3.7–4.0 (Van Schmus, 1995). This seems to indicate that metasomatic activities (caused by primary dissolution and by secondary mineral formation) had occurred. The main factors that influence the mineralogical structure of a metamorphic rock are the bulk composition of the rock, the pressure and temperature conditions at the time of crystallization and the composition of the fluid phase in the rock during metamorphism. The bulk composition of the metamorphic rock may be inherited from the original sedimentary or igneous precursor, or may be chemically altered by metasomatic activity during the metamorphic process. The average Th/U ratio for the BF samples was 3.27. This indicates that the rock have undergone alteration, weathering or other metasomatic activities. The samples from Ashiaman had very high Th/U ratio of 19.34.. In this case, it can be explained that the radioelements have been quite fractionated during magmatic differentiation, weathering or other metasomatic activities and U has been enriched at a lower level relative to Th. Hence, ²³²Th activity concentrations are generally higher than ²³⁸U activity concentrations.

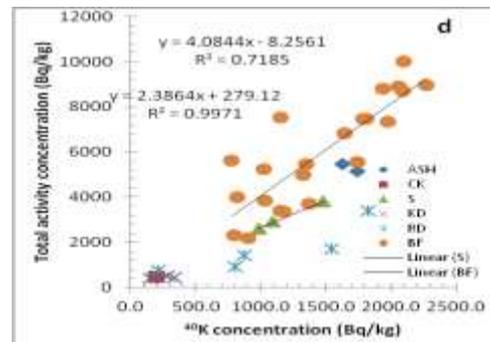
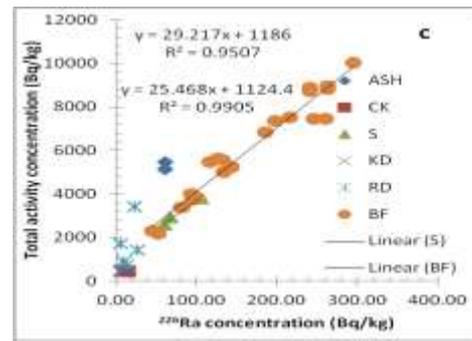
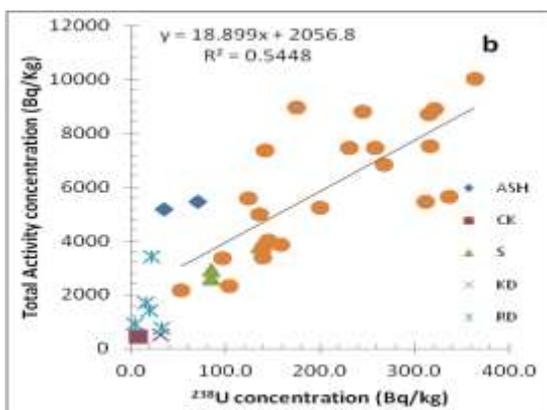
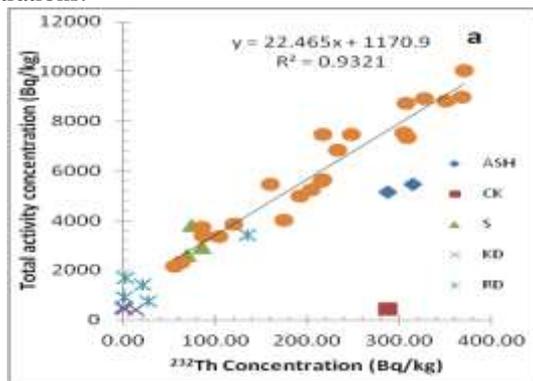
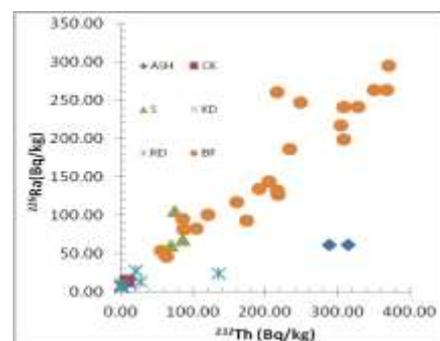
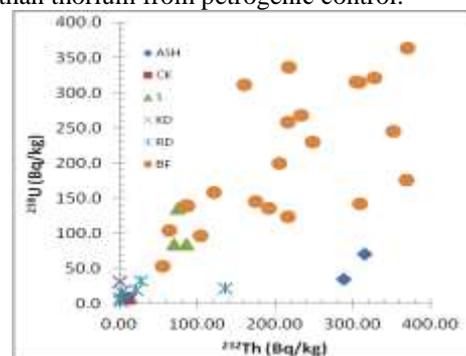


Fig.3 (a-d) The total activity concentration vs individual radionuclide activity concentration

Another possible explanation for the erratic behavior of U is its presumed high mobility in oxidized form, which might permit its selective removal from magma in volatile phases ([Rogers and Ragland, 1961). Uranium can be oxidized to the hexavalent state, but thorium, cannot exist in this valence state. Hexavalent uranium is considerably more soluble than tetravalent thorium and furthermore, forms many very soluble compounds. If the uranium is oxidized during magmatic activity, then it will tend to enter volatile phases and escape from the magma. The presence of ferric iron in igneous rocks (magnetite) indicates that oxidizing capacity is present in magmas. The postulated loss of uranium from the magma explains the irregular distribution of uranium and its generally wider deviation than thorium from petrogenetic control.



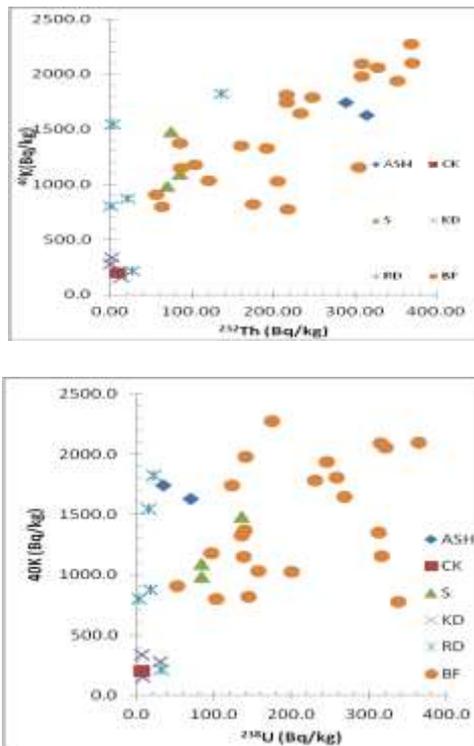


Fig. 4 (a-d) Variation diagrams of the radionuclides

The K/Th ratio was 0.63, which is close to the typical values obtained in a large variety of unaltered lithologies (Chiozzi et al., 2002; Galbraith and Saunders, 1983), but it should be mentioned that these values are highly variable and depend on the geological origin of the sample (Roger and Adams, 1969; Tzortzis and Tsertos, 2004; Chiozzi et al., 2002).

Calculation of radiological effects

The main objective of measuring radioactivity is to make an estimate of radiation doses likely to be delivered externally to the general public. The contribution of natural radionuclides to the absorbed dose rate in air depends on the concentrations of the radionuclides in the rock and soil. The absorbed gamma dose rates in air at one (1) metre above the ground surface for the uniform distribution of radionuclides (²³⁸U, ²³²Th and ⁴⁰K) were calculated using the formula proposed by UNSCEAR (2000)

$$\dot{D} = \sum x A_x C_x \tag{4}$$

where A_x (Bq/kg) is the activities of the radionuclides and C_x (nGy/h per Bq/kg) their corresponding dose conversion factors. The dose conversion factors reported by UNSECAR (2000) was used assuming that contribution from other naturally occurring radionuclides were insignificant. Hence the absorbed dose rate was calculated using the equation below

$$D(nGy/h) = 0.462C_U + 0.604C_{Th} + 0.0417C_K \tag{5}$$

where C_U , C_{Th} and C_K are the activity concentrations of U, Th and K respectively.

The calculated absorbed dose rates are presented in Table 3. The absorbed dose rates varied from 96.00 to 479.26 nGy/h for the BF samples, 262.84 to 290.59 nGy/h for the ASH samples, 122.24 to 169.17 nGy/h for the shale soil samples, 18.27 to 26.62 nGy/h for the KD samples and from 35.68 to 168.18nGy/h for the RD samples. According to UNSCEAR (2000) report, the dose rate in air outdoors from terrestrial gamma-ray in normal circumstances is about 57nGy/h. Thus the absorbed dose rate values for the samples were higher except for the KD samples. The largest contributor to the absorbed dose in air was from ²³²Th. Figure 5 illustrates the measured relative contribution to

the total dose outdoors for the BF samples which had the highest dose rates.

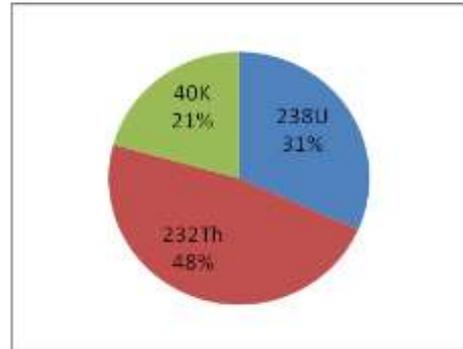


Fig.5. Relative contribution to absorbed dose rate in air outdoors for the BF samples

The annual effective dose equivalent (AEDE) was calculated from the absorbed gamma dose rate using the conversion coefficient from absorbed dose in air to the effective dose (0.7Sv/Gy) and the outdoor occupancy factor (0.2). Thus the AEDE was calculated using the formula

$$\begin{aligned} \dot{E} (mSv/yr) &= \dot{D}(nGy/h) \times 8760 \text{ h.yr}^{-1} \times 0.7 \times (10^3 mSv/10^9) nGy \times 0.2 \\ &= \dot{D} \times 1.23 \times 10^{-3} (mSv/yr) \end{aligned} \tag{6}$$

The AEDE values for the samples were high (table 3) as the worldwide average annual effective dose is approximately 70Sμv/y (UNSCEAR, 2000). Despite the high AEDE values, the Accra Plains can be classified as a low-level natural radiation area (LLNRA) in accordance with the classification scheme proposed by Sohrabi (1998). According to the classification scheme, a LLNRA is an area or a complex of dwelling where potential public exposure fails below or equal to twice the average global annual effective dose from natural sources, i.e. by UNSCEAR report 1993, $2 \times 2.4 = 4.8mSv/y$ or $\approx 5mSv/y$.

In areas with the normal background radiation, the average annual external effective dose rate from the terrestrial radionuclides is 0.46 mSv/year(UNSCEAR, 1993; Bennett, 1997; Mohanty et al.,2004).

The gamma ray radiation hazards due to the specified radionuclides were assessed by two different indices, the radium-equivalent activity and external radiation hazard. The radium equivalent activity (Ra_{eq}) is defined by the estimate that 1Bq/kg of ²²⁶Ra, 0.7Bq/kg of ²³²Th and 13 Bq/kg of ⁴⁰K produce the same gamma ray dose and mathematically represented by $Ra_{eq}(Bq/kg) = A_{Ra} + 1.43A_{Th} + 0.077A_K$

Where A_{Ra} , A_{Th} and A_K are the activity concentrations of ²²⁶Ra, ²³²Th and ⁴⁰K respectively. The permissible value of Ra_{eq} is ≤ 370 Bq/kg (Galy et al, 2008). The mean calculate Ra_{eq} values for Kwabenya (BF) and Ashiaman samples were higher than the permissible values. The BF values increased with depth indicating increase in the risk hazard with depth.

The external hazard index (H_{ex}) was used to measure the external hazard due to the emitted gamma radiation. It was calculated using the equation below

$$H_{ex} = C_{Ra}/370 + C_{Th}/259 + C_K/4810 \leq 1 \tag{10}$$

The BF samples had high external hazard indices which increase with increase in depth. The samples from Ashiaman also had external hazard indices greater than one (1).

Conclusion

The activity concentration of ²³⁸U, ²³²Th, ²²⁶Ra, ⁴⁰K and ¹³⁷Cs were measured in 38 rock and soil samples collected from

6 different locations in the Accra Plain. A high resolution gamma ray spectrometer was used to carry out the measurement. The concentration of the radionuclides increases with petrogenic evolution and the relative enrichment is $K > Th > U > Ra$. The median activity of ^{238}U , ^{232}Th , and ^{40}K for the samples were higher than worldwide median value of rocks (UNSCEAR, 2000) except for the activity concentrations for the samples from around the Krobo Mountains and Tema. The activity concentration of ^{40}K was found to be higher than the other radionuclides. The high ^{40}K concentration may be due to high potassium feldspar mineral contents in the samples. Minimum concentrations of ^{137}Cs were only found in the samples from Ashiaman, Okwenya as well as in one of the rock samples from the foot of the Krobo Mountains.

The possible loss of uranium during the later stages of rock differentiation may result from oxidation of uranium to the uranyl ion during the evolution of suites of igneous rocks. This conclusion is based on the assumed greater solubility of uranyl ion than of tetravalent uranium or thorium in the volatile igneous phase. It is however possible that fractionation occurs between the tetravalent U and Th ions, for the two ions are not identical, though they are quite similar. The presence of ferric iron in igneous rocks (magnetite) indicates that oxidizing capacity is present in magmas.

The Th/U ratio indicated that the radioelements in the rocks samples from Tema have not been significantly fractionated during weathering or involved in metasomatic activity whilst those from BNARI and the Krobo Mountain areas had undergone alteration, weathering or other metasomatic activities.

The median absorbed dose rate values for the samples had higher values than the worldwide value of 51nGy/h except for the dose rate calculate for the samples from the Krobo Mountains area. The ^{232}Th series gave the largest contribution to the absorbed dose in air outdoors.

The gamma ray radiation hazards assessed using the radium-equivalent activity and external radiation hazard indicated that risk levels in BF and Ashiaman were high as they had values exceeding the recommended limit.

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Table 1: Descriptive statistics of the activity concentration

	Minimum	Maximum	Mean	Median	Std Deviation
U ppm	0.28	29.48	11.57	10.98	9.08
Th ppm	0.11	91.01	39.33	36.35	30.02
Ra ppm	0.45	23.93	9.21	7.63	7.36
K %	0.5	7.36	4.08	4.05	1.93

Table 2: Abundances of radionuclides in the geological formation

Samples	²³⁸ U(Bq/Kg)	²³² Th(Bq/Kg)	²²⁶ Ra(Bq/Kg)	⁴⁰ K(Bq/kg)
BF	53.1 - 363.5	55.9 - 370.43	45.83 - 295.01	776.0 - 2274.3
KD	BDL - 31.8	BDL - 13.51	4.04 - 10.11	145 - 377.4
RD	BDL - 32	1.09 - 135.74	5.91 - 27.06	214.6 - 1824.1
ASH	34.7 - 70.6	288.2 - 314.69	60.93	1628.4 - 1744.2
CK	7.6	9.07	14.28	199.3
S	84.3 - 135.5	69.7 - 85.32	59.91 - 105.47	983.1- 1478.4

BDL-below detection limit

Table 3. Absorbed dose rate (D), Annual effective dose equivalent (AEDE), External hazard index (H_{ex}) and Radium equivalent activity (Ra_{eq}) for the rock and soil samples

ID	Absorbed dose rate (D) nGy/h	Annual Effective Dose equivalent (AEDE) mSv/y	External hazard index (Hex)	Radium equivalent activity Ra _{eq} Bq/kg
ASH1	290.59	0.36	1.72	636.23
ASH2	262.84	0.32	1.64	607.39
CK1	17.28	0.02	0.11	42.6
S1	136.01	0.17	0.74	274.02
S3	122.24	0.15	0.63	235.27
S4	169.17	0.21	0.88	325.62
KD2	26.62	0.03	0.08	29.18
KD3	18.27	0.02	0.09	33.61
KD4	18.59	0.02	0.11	41.33
RD1B	168.18	0.21	0.97	357.86
RD2A	-	-	0.76	281.97
RD2C	40.05	0.05	0.18	68.05
RD3B	35.68	0.04	0.2	73.03
RD3C	58.06	0.07	0.34	124.81
RD4B	73.53	0.09	0.35	128.64
BF72	406.32	0.50	2.47	914.73
BF69	479.26	0.59	2.66	986.43
BF66	431.95	0.53	2.34	867.99
BF63	398.25	0.49	2.6	964.45
BF60	334.07	0.41	2.14	791.34
BF57	418.85	0.51	2.28	842.98
BF54	330.7	0.41	1.99	738.68
BF51	378.04	0.46	2.00	740.93
BF48	325.67	0.40	1.92	709.47
BF45	333.15	0.41	1.74	645.81
BF42	296.95	0.36	1.21	449.49
BF39	233.66	0.29	1.38	510.3
BF36	189.01	0.23	0.95	352.7
BF33	96.00	0.12	0.55	202.8
BF30	319.3	0.39	1.34	497.68
BF27	260.47	0.32	1.55	575.98
BF24	164.6	0.20	0.79	294.31
BF21	258.73	0.32	1.39	515.93
BF18	119.48	0.15	0.53	198.02
BF15	173.39	0.21	0.87	323.06
BF12	156.55	0.19	0.87	321.28
BF9	206.52	0.25	1.09	405.3