

Assessment of Natural Radioactivity in Soil and Water Samples from Aden Governorate South Of Yemen Region

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Abstract: The level of natural radioactivity in soils and water of 74 samples collected from locations at Aden governorate south of Yemen was measured. Concentrations of radionuclides in soils and water samples were determined by gamma-ray spectrometer using a NaI (TI) detector with specially designed shield. The mean activity level of the natural radionuclides ²²⁶Ra, ²³²Th and ⁴⁰K is 30.41 ± 2.4 , 36.26 ± 2.9 and 358.12 ± 26.54 Bq kg⁻¹, respectively. These values are well within values reported elsewhere in the country and in other countries with similar environments. The study also examined some radiation hazard indices. The mean values obtained are, 215 ± 37 Bq kg⁻¹, 50.10 ± 6 nGy h⁻¹, 0.34 mSv y⁻¹, 0.30 and 0.34 for Radium Equivalent Activity (Ra_{eq}), Absorbed Dose Rates (D), Annual Effective Dose Rates (Eff Dose), External Hazard Index (H_{ex}) and Internal Hazard Index (H_{in}) respectively. All the health hazard indices are well below their recommended limits. The paper recommends further studies to estimate internal and external doses from other suspected radiological sources to the population in Aden governorate.

Keywords: Absorbed dose, south Yemen, Aden governorate, Water, Soil, Natural Radioactivity.

1. INTRODUCTION

Natural environmental radioactivity and the associated external exposure due to gamma radiation depend mainly on the local geological and geographical conditions and appear at different levels in each region in the World (UNSCEAR, 2000). The natural terrestrial gamma dose rate is an important contributor to the average dose received by the world's population (Tso and Leung, 2000; Senthilkumar et al., 2010). Estimation of the radiation dose distribution is vital in assessing the health risk to a population and serves as a reference for documenting changes in environmental radioactivity due to anthropogenic activities (Obed et al., 2005). Monitoring for radioactive materials are therefore of primary importance for humans, organisms and for environmental protection, but rapid and accurate methods for the assay of radioactivity is essential (El-Bahi, 2004). When outdoors, humans are exposed to natural terrestrial radiation that originates predominantly from the upper 30 cm of the soil (Chikasawa et al., 2001). Humans are also exposed by contamination of the food chain which occurs as a result of direct deposition of radionuclides on plant leaves, root uptake from contaminated soil, sediment or water (Arogunjo et al., 2004), and from direct ingestion of contaminated water (Avwiri and Agbalagba, 2007). To assess these exposures, radioactivity studies have been previously carried out in soil and sediment samples in other parts of the world, some similar to those in south Yemen (e.g. Selvasekarapandian et al., 2005; Kannan et al., 2002; Kirchner et al., 2002; Al-Junda et al., 2003; Avwiri et al., 2005; A.I.; Obed et al., 2005; Patra et al., 2006; Khaled, S. I. A., 2006; Diab et al., 2008; Senthilkumar et al., 2010; Abd El-mageed, 2010 Abd El-Hadi El-Kamel et al., 2012;) and in water samples (Onoja, 2004; Avwiri et al., 2005, 2007; Avwiri and Agbalagba, 2007 and Shaban Harb et al., 2012). This present study assesses the specific activities and examines some of the radiation hazard

indices of these naturally occurring radionuclides (^{226}Ra , ^{232}Th and ^{40}K) in soil and water samples from Aden governorate south of Yemen, using gamma-ray spectrometry. The values reported in this study constitute a baseline for radioactivity in the study area as no such study has been carried out before.

2. MATERIALS AND METHODS

2.1. Description of study area

The study was conducted on an area consisted of 80 hectares cultivated farms soil in the Aden governorate 20km from the famous city of Aden (called Capital of south Yemen) in the Aden governorate south of Yemen. The location of the area is $12^{\circ}54'.211\text{N}$ and $12^{\circ}53'.294\text{E}$ and at the sea level. The annual rainfall in this area is 100mm. The only source of irrigation is tube-wells having EC of 4.8 ds/m; sodium absorption ratio (SAR) is equal to 39.5 and sodium carbonate (RSC) ranging 18-23. The soil is medium texture with salinity ranging 21-6 desi semin/meter (ds/m) from 1m depth of soil profile (Sakran, Sh. 1993).

2.2. Sampling and samples preparation

2.2.1. Soil sampling

In order to measure the natural radioactivity in the cultivated farms soil samples have been taken from the region of Aden governorate south of Yemen. In this area, there are several species of green vegetables which are planted such as potatoes, sweet pepper, tomatoes, cauliflower and of large surfaces of vegetables. Most of the cultivated areas considered as the main source of vegetables for citizens who live in Aden governorate. For a complete survey the natural radioactivity of irrigation water was measured as well (Baykara and Dogru, 2006). The soil under investigation is an argillaceous soil contains 60% clay, 24% loam and 16% sand. The soil textures for all samples were very similar as they are taken from the same region. The soil is slightly alkaline (pH 8.5). Each soil sample was a composite collected in a black polythene bags from five sample locations, each of about 200 cm^2 , within an area of approximately 100 m^2 . The samples were collected to a depth of 10-15 cm which represents the soil depth variation given the purpose of the study and the soil characteristics (soil permeability). After removing the stones and inorganic materials, the samples were dried in an oven at about 100°C then crushed and sieved through 1 mm mesh sieve. The sieved portion of the samples was transferred into 500 ml Marinelli beakers for gamma spectrometry and sealed for four weeks to reach secular equilibrium between the thorium and radium contents of the sample and their daughter radionuclide's.

2.2.2. Water Samples

Because no running water in this area 36 groundwater samples were collected from the study area that used as irrigation water. Measuring pH values as well as conductivity for water samples were measured in the laboratory. Standard polyethylene Marinelli beakers (1.4 liter) were used as a sampling and measuring container. Before use, the containers were washed with dilute hydrochloric acid and rinsed with distilled water. Each beaker was filled up to brim and a tight cap was pressed on so that the air was completely removed from it. The collected water samples were left for an overnight period in polyethylene containers to allow setting of any suspended solid materials and for each samples a clear supernatant was separated decantation. The clear solution was acidified by adding 0.5 ml of conc. HNO_3 per liter, to prevent any loss of radium isotopes around the container walls, and to avoid growth of microorganisms. The water samples were then homogenized well by shaking. The final acidity of water samples reaches pH-2. The samples were stored for over 30 days to reach secular equilibrium before radiometric analysis (Onoja, 2010).

2.3. Experimental setup

Each sample was measured with a gamma-ray spectrometer consisting of a NaI (Tl) setup and multichannel analyzer 8192 channel, with the following specifications: resolution (FWHM) at $1.33\text{MeV } ^{60}\text{Co}$ is 60keV, relative efficiency at $1.33\text{MeV } ^{60}\text{Co}$ is 7.5 %. The detector is shielded in a chamber of two layers starting with stainless steel (10 mm thick) and lead (30 mm thick). The sample was placed over the detector for at least 10 h. The spectra were either evaluated with the computer software program Maestro (EG&G ORTEC), or manually with the use of a spread sheet (Microsoft Excel) to calculate the natural radioactivity. ^{226}Ra activity of the samples was determined via its daughters (^{214}Pb and ^{214}Bi) through the intensity of the 295.22, 351.93keV, for ^{214}Pb Gamma-lines and 609.31, 1120, 1764.49keV, for ^{214}Bi Gamma-lines. ^{232}Th activity of the sample was determined from the daughters (^{228}Ac), (^{212}Pb) and (^{208}Ti) through the intensity of 209.25, 338.32, 911.2keV Gamma-lines for (^{228}Ac), (^{212}Pb) emissions at 238.63keV and (^{208}Ti) emissions at 583.19,

2614keV Gamma-lines. ⁴⁰K activity determined from the 1460.7keV emissions Gamma-lines. The Minimum Detectable Activity (MDA) for, ²²⁶Ra, ²³²Th and ⁴⁰K in soil and water were determined and are given in Table 1 and Table 2. The MDA in groundwater for the radionuclide ⁴⁰K is 1.942Bq/l. The gamma ray spectrum was accumulated for up to 86400 s (24 h) for each sample. In our case, inactive soil and sterilized water were used as blank and the time of collection was 194400 s (54 h). The relative efficiency curve of the detector was determined from soil samples spiked with a multinuclide standard solution (QCYB41) DKD (Germany) (E.O. Agbalagba, 2011).

Table 1: Detection limit and minimum detectable activity table for soil samples (Bq/kg).

Radionuclide	Energy (keV)	Critical level(Lc) (counts)	Detection limit (Ld) (counts)	Minimum detectable activity (MDA) (Bq)
²¹⁴ Bi	609.3	40.05	83.03	0.553
²¹⁴ Bi	1120.2	64.38	131.82	3.42
²¹⁴ Bi	1764.5	38.33	79.58	2.51
²¹² Pb	238.6	29.71	62.29	0.854
²²⁸ Ac	911.2	37.05	77.01	0.368
⁴⁰ K	1460.8	124.33	252.06	10.01

Table 2: Detection limit and minimum detectable activity table for water samples (Bq/l).

Radionuclide	Energy (keV)	Critical level(Lc) (counts)	Detection limit (Ld) (counts)	Minimum detectable activity (MDA) (Bq)
²¹⁴ Bi	609.3	51.77	106.54	0.074
²¹⁴ Bi	1120.2	38.54	80.01	0.237
²¹⁴ Bi	1764.5	55.29	113.59	0.399
²¹² Pb	238.6	30.16	63.19	0.093
²²⁸ Ac	911.2	37.05	77.01	0.035
⁴⁰ K	1460.8	188.40	380.57	1.942

3. RADIATION HAZARD INDICES CALCULATION

It is justifiable to exploit as many as possible of the known radiation health hazard indices to arrive at a safe conclusion on the health status of an exposed person or environment. The indices estimated in this report are as follows. To represent the activity levels of ²²⁶Ra, ²³²Th and ⁴⁰K by a single quantity, which takes into account the radiation hazards associated with each component, a common radiological index has been introduced (Diab et al., 2008). This index is called the Radium Equivalent activity ($R_{a_{eq}}$) and is mathematically defined by (UNSCEAR, 2000):

$$R_{a_{eq}} \text{ (Bq.Kg}^{-1}\text{)} = (1.43 \times A_{Th}) + A_{Ra} + (A_K \times 0.077) \quad (1)$$

Where A_{Ra} , A_{Th} and A_K are the activity concentrations of ²²⁶Ra, ²³²Th and ⁴⁰K respectively. In the above relation, it has been assumed that 10Bq kg⁻¹ of ²²⁶Ra, 7Bq kg⁻¹ of ²³²Th and 130Bq kg⁻¹ of ⁴⁰K produce equal gamma dose. The absorbed dose rates (D) due to gamma radiation in air at 1m above the ground surface, assuming uniform distribution of the naturally occurring radionuclides (²²⁶Ra, ²³²Th and ⁴⁰K) have been calculated based on guidelines provided by

UNSCEAR (2000). It has been assumed that the contributions from other naturally occurring radionuclides were insignificant. Therefore, D can be calculated according to UNSCEAR (2000) as:

$$D \text{ (nGy.h}^{-1}\text{)} = 0.462A_{\text{Ra}} + 0.621A_{\text{Th}} + 0.0417A_{\text{K}} \quad (2)$$

To estimate annual effective dose rates, the conversion coefficient from absorbed dose in air to effective dose (0.7SvGy⁻¹) and an outdoor occupancy factor (0.2) proposed by UNSCEAR, 2000 are used. Therefore, the annual effective dose rate (mSvy⁻¹) was calculated by the formula (UNSCEAR, 2000):

$$\text{Effective dose rate (m Sv y}^{-1}\text{)} = D \text{ (nGy h}^{-1}\text{)} \times 8760 \text{ h} \times 0.8 \times 0.7 \text{ SvGy}^{-1} \times 10^{-6} \quad (3)$$

A widely used hazard index (reflecting external exposure) called the external hazard index H_{ex} is defined as follows (UNSCEAR, 2000):

$$H_{\text{ex}} = (A_{\text{Ra}}/370 + A_{\text{Th}}/259 + A_{\text{K}}/4810) \leq 1 \quad (4)$$

In addition to the external hazard index, radon and its short lived progeny are also hazardous to the respiratory organs. The internal exposure to radon and its daughter progenies is quantified by the internal hazard index H_{in} (UNSCEAR, 2000), which is given by the equation:

$$H_{\text{in}} = (A_{\text{Ra}}/185 + A_{\text{Th}}/259 + A_{\text{K}}/4810) \leq 1 \quad (5)$$

The values of the indices (H_{ex}, H_{in}) must be less than unity for the radiation hazard to be negligible (Diab et al., 2008).

4. RESULTS AND DISCUSSION

From the gamma spectrometric analysis, three naturally occurring radionuclides were determined (²²⁶Ra, ²³²Th and ⁴⁰K) in the soil and water samples. Table 2 presents the results of activities concentrations of the soil samples. Radium-226 ranged from 15.39Bqkg⁻¹ in Beer Ahmed to 108.18Bqkg⁻¹ in Al-Masabian area with mean value 59.39±4.76Bqkg⁻¹, While ²³²Th ranged from 30.26Bq kg⁻¹ in Beer Ahmed to 132.12Bq kg⁻¹ also in Al-Masabian area with mean value 71.32±5.71. The activity concentration of ⁴⁰K was observed to be comparatively higher than that of both ²²⁶Ra and ²³²Th in all the soil-sampling locations studied. Its value ranged from 289.5Bq kg⁻¹ in Beer Fadle area to 1120.23 Bq kg⁻¹ in Al-Masabian area with mean value 697. 9±51.47Bq kg⁻¹ (Table 2).

Table 3 presents the results of the activity concentrations in water samples. They ranged from 0.22 to 2.67 Bql⁻¹with a mean value of 1.44±0.072Bql⁻¹for ²²⁶Ra. Activity concentrations for ²³²Th ranged from 0.15 to 3.06 Bql⁻¹with a mean value of 1.2±0.074 Bql⁻¹, while ⁴⁰K activity concentrations ranged from 7.87 to 26.02 Bql⁻¹with a mean value of 18.34±1.61Bql⁻¹.

Table 2 Activity concentrations of natural radionuclides (Bq kg⁻¹) in soil samples from the Aden Governorate South of Yemen Region.

Location	Sample No.	²²⁶ Ra		²³² Th		⁴⁰ K	
		Mean	Range	Mean	Range	Mean	Range
Beer Ahmed	9	49.9±3.98	15.39-88.05	59.53±4.79	30.26-98.70	642.57±48.46	375.50-895.9
Beer Fadle	9	46.11±3.77	31.66-73.68	56.29±4.5	36.56-89.12	605.96±38.49	289.5-781.51
Daar-saad	9	65.34±5.22	35.73-104.43	79.89±6.38	41.86-129.48	723.28±54.49	580.90-967.2
Al-Masabian	9	76.21±6.10	52.83-108.18	89.60±7.16	65.22-132.12	819.79±64.46	621.78-1120.23
Mean Value		59.39±4.76		71.32±5.71		697. 9±51.47	

Table 3 Activity concentrations of natural radionuclides (Bq L⁻¹) in water samples from the Aden Governorate South of Yemen Region.

Location	Sample Number (n)	²²⁶ Ra		²³² Th		⁴⁰ K	
		Mean	Range	Mean	Range	Mean	Range
Beer Ahmed	9	1.48±0.073	(0.33-2.67)	1.25±0.076	(0.15- 2.72)	A. 17.0 5±1.46	(7.87-19.48)
Beer Fadle	9	1.592±0.078	(0.26-2.45)	1.22±0.059	(0.32-2.31)	16.93±1.45	(13.07-21.32)
Daar-saad	9	1.081±0.053	(0.22-2.16)	1.1±0.065	(0.39-1.67)	19.98±1.72	(15.05-26.02)
Al-Masabian	9	1.73±0.086	(1.08-2.29)	1.58±0.096	(0.57-3.06)	21.15±1.82	(15.55-23.97)
Mean Value		1.44±0.072		1.2±0.074		18.34±1.61	

The overall results show that the ²²⁶Ra, ²³²Th and ⁴⁰K are not uniformly distributed in soil, but the radioactivity varies, often greatly, over a distance of some meters. The results obtained in soil samples are slightly lower than some values reported in similar environments in other country (Kannan et al., 2002; Al-Junda et al., 2003 and Patra et al., 2006). However, all these values are well within the worldwide average concentration of these natural radionuclides in soil as reported by UNSCEAR (2000), and they are below the activity values reported by Obed et al., 2005; Diab et al., 2008 and Senthilkumar et al., 2010. The use of fertilizers in large extent have affected radionuclides concentration, especially potassium containing fertilizers are the one of the cause of presence of high activity of ⁴⁰K in soil (Bhatti, et al., 1994). The level of ²³²Th observed may be due to the geology of the area (including the presence of metamorphic rocks like shale). However, a detailed geochemical investigation is required to reach a definite conclusion on causes.

The radium equivalent activities due to the presence of naturally occurring terrestrial radionuclides ²²⁶Ra, ²³²Th and ⁴⁰K were calculated based on their activities in soils determined by gamma-ray spectrometry and using equation (1). The Ra_{eq} due to radionuclides varied from 173±29 to 267±46Bqkg⁻¹ for soil with overall mean value of 215±37Bqkg⁻¹, which is 42% less than the 370 Bqkg⁻¹ recommended maximum levels of radium equivalents in soil (Shaban Harb, 2012), Thus, the soil are suitable for use as building materials.

The external gamma dose rates were calculated based on their activities in soil and water samples using equation (2). The dose rates generally varied from 1.94 to 121.40mGy h⁻¹ with a mean value of 50.10mGy h⁻¹ which is within the range of values given in UNSCEAR (2000) report {57 (18-93) mGy h⁻¹}. The outdoor annual effective doses ranged from 0.009 to 0.83mSvyr⁻¹ with a mean value of 0.34mSvyr⁻¹ as calculated using equation (3), while the worldwide average annual effective dose is approximately 0.5 mSv yr⁻¹ and the results for individual countries being generally within the 0.3-0.6 mSv yr⁻¹ range (UNSCEAR, 2000; Senthilkumar et al., 2010).

Thus, these results are less than the average worldwide limits as reported by UNSCEAR (2000). The external hazard index H_{ex} was calculated using equation (4). Its values ranged from 0.011 to 0.72 with a mean of 0.3.

The internal hazard index H_{in} was calculated using equation (5), the value of H_{in} ranged from 0.013 to 0.93 with a mean value of 0.34. The values of H_{ex} and H_{in} of all samples studied in this work are far less than unity.

Table 4 Radiation hazard indices of soil and water samples from the Aden Governorate South of Yemen Region.

Location	Sample type	Radiation hazard indices				
		Ra _{eq} (Bq/kg)	D (nGy h ⁻¹)	E _{ff} Dose (m Sv y ⁻¹)	H _{ex}	H _{in}
Beer Ahmed	Soil	185. ±32	84.67±13	0.58	0.50	0.63
	Water	Nil	2.16±0.25	0.010	0.012	0.016
Beer Fadle	Soil	173±29	79.28±9	0.54	0.47	0.59
	Water	Nil	2.18±0.25	0.011	0.012	0.017
Daar-saad	Soil	235±40	106.73±12	0.73	0.64	0.81
	Water	Nil	1.94±0.23	0.009	0.011	0.014
Al-Masabian	Soil	267±46	121.40±14	0.83	0.72	0.93
	Water	Nil	2.48±0.29	0.012	0.014	0.018
Mean Value		215±37	50.10±6	0.34	0.30	0.34

5. CONCLUSION

Samples of waters and agricultural soils of the studied region were measured for their radioactivity content using Gamma spectrometry technique. These data show that the activity concentration of naturally occurring radionuclides in soil samples were within the world average ranges which are 35(10-50), 35(7-50) and 370(100-700) Bq/kg for ²²⁶Ra, ²³²Th and ⁴⁰K respectively (UNSCEAR, 2000). However, the water samples show higher concentrations than the soil ones. The variations in the activity levels have been observed to be lying within the activity values measured all over the world, especially for soil samples but not water samples. This work has established baseline information on the natural radioactivity status of four areas in Aden governorate south of Yemen, which will serve as a reference for future studies. We recommend that normal occupational health and environmental protection measures designed for non-radiological hazards in the environment are sufficient to protect against any future natural radiological hazard that may arise in the area. However, further study may be necessary to estimate the external and internal doses from other suspected radiological sources within the Aden governorate

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