Enhancement of natural radioactivity in farm surface soils from Abyan Delta in Yemen

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Abstract: Radium, thorium and potassium analyses have been made in farm soil samples collected from Abyan Delta in Yemen using gamma ray spectrometry. Abyan Delta is an agricultural area that produces many agricultural crops in Yemen. The results show that ^{226}Ra, ^{232}Th and ^{40}K radionuclides were present in average concentrations of 34.9 ± 2.9, 84.5 ± 7.8, 1232.7 ± 40 Bq kg^{-1} for farm surface soil irrigation by flood water. The corresponding values for farm surface soil irrigation by ground water were 30.24 ± 2.7, 63.64 ± 5.7, 1096 ± 36 Bq kg^{-1}, respectively. The radium equivalent activity (Ra_{eq}), the external hazard index (H_{ex}), the internal hazard index (H_{in}), and absorbed dose in air were calculated. The data were discussed and compared with those given in the literature. Chemical analysis for elemental oxides was also carried out along with the measurement of pH of soil samples.

Keywords: farm soil; natural radioactivity; absorbed dose rate; Abyan Delta.

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1 Introduction

The knowledge of the concentrations and distributions of the radionuclides in soil is of interest since it provides useful information in the monitoring of environmental contamination by natural radioactivity. The concentrations of natural radioactivity such as radioactive series (^{238}U and ^{232}Th) and radioactive isotope ^{40}K and the related external exposure due to gamma radiation depend mainly on the geological and geographical conditions. Higher radioactivity in soil samples may be linked to the contribution of the parent materials that constitute the soil type. For instance, soil derived from granite will have a higher radioactivity than the soil derived from the other rock types (Malanca et al., 1996; Ramli, 1997).

Agriculture soil contamination is probably the most relevant event because radionuclides could reach humans via the food chain. Soil contamination by radionuclides and possible countermeasures have been recently reviewed by Zho and Shaw (2000). On average, 79% of the radiation to which humans are exposed is from natural sources, 19% is from medical applications, and the remaining 2% is from fallout of weapons tests and the nuclear power industry (Wild, 1993).

Very large amounts of ^{238}U and ^{232}Th series radionuclides have been released to the marine environment during (or after) nuclear weapons tests or the nuclear power industry, and during several no nuclear industrial processes: Technologically Enhanced, Naturally Occurring Radioactive Material (TENORM) such as phosphogypsum phosphate ore processing (Absi et al., 2004) gas and oil production (El Mamoney and Khater, 2004), uranium mining or sewage treatment, used of coal, oil, and natural gas to produce electric energy (Bolivar et al., 1995). These activities can enhance the local activity concentrations.

The dynamics of soil water, as well as the texture and structure of soil, have a direct impact on radionuclide speciation. Chemically unchanged substances can be partially transferred through water flow, whereas slow infiltration favours interaction with the soil matrix and soil solution (Wild, 1993). The other factor also affecting the behaviour of radionuclides in soil is rainfall amounts (Koch-Steindl and Pröhl, 2001). Rainwater helps to remove radioactivity from the troposphere and transport it to the Earth's surface.

There are few studies carried out in Yemen on natural radioactivity measurements in soil (El-Mageed et al., 2010; El-Kamel et al., 2012). The interest behind the study in this part of Yemen is mainly due to its contribution to food production. This paper is the first research representing the natural radioactivity study in the agricultural soil samples collected from Abyan Delta, Yemen, measured by using gamma spectrometry technique. The radiation health hazard indices were also calculated and are presented in this paper.

2 Materials and methods

2.1 Study area

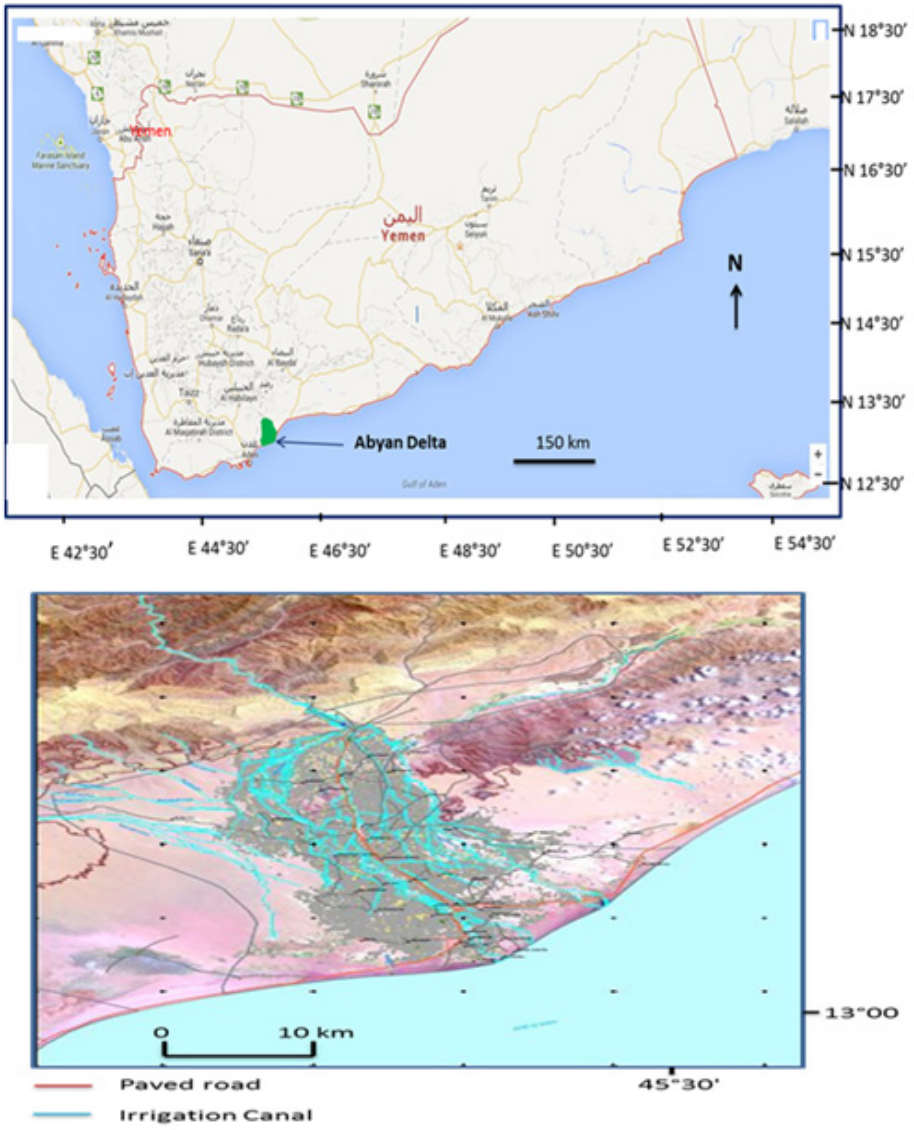
Abyan Governorate is located at the southern part of Yemen. It extends for about 280 km along the Gulf of Aden coast with inland varying depth from 30 to 70 km, the study areas shown in Figure 1. Abyan Governorate comprises three main basins: Wadi Bana, Wadi Hassan and Wadi Ahwar. The former wadis, Bana and Hassan, have large catchment areas of 7400 and 3200 km^2, respectively. The run-off accumulating at the lower end of both wadis is drained to the combined Bana and Hassan Delta and designated as the Abyan Delta. Abyan Delta is a vast plain with its highest point in Bateis (around 170 m above sea level), where Wadi Bana enters. The distance from the apex of the delta to the Gulf of Aden is approximately 30 km, while the base of the delta is about 20-km long. It is bordered in the north and the north-east by high mountain chains (from 700 to 1000 m) (Ewea, 2007).

The basement rocks exposed in the northern part of the study area are composed of mid to upper proterozoic metamorphic and intrusive rocks of the southern part of the Arabian Shield. The metamorphic rocks include metavolcanics, amphibole-biotite schists and gneisses, and higher grade migmatites. The intrusive rocks include granites and gabbros (KOMEX, 2002).

The large catchment areas of wadis Bana and Hassan extend upward and drain high mountainous ground. However, the direct annual rainfall on Abyan Delta is very low and hardly goes beyond the 100 mm per year. Mean annual rainfall of wadis Bana and Hassan are 370 and 250 mm, respectively. Such depth of rainfall constitutes annual

volume of water of 2,738,800 million m^3, respectively. Although both wadis have a large catchment extending upward to the areas of high rainfall, rainfall is insufficient for rain-fed agriculture in the delta of Abyan (Atkins and Binnie, 1984).

Figure 1 Base map of the Abyan Delta, Republic of Yemen



Major floods usually occur in both summer and autumn seasons. Along with irrigation by flood water, also groundwater (abstraction by hand dug wells and tube wells) has been used for irrigation in Abyan Delta.

In this experimental work, we classified the soil samples into two broad groups depending on the type of irrigation water:

1 Soil samples from the farmland irrigation by flood water (70 samples).

2 Soil samples from the farmland irrigation by groundwater (40 samples).

2.2 Sampling and sample preparation

A total of 110 surface soils, at 5-cm depth, were collected using a stainless steel sampler. After removing the stones and inorganic materials, the samples were air-dried and dried in an oven at about 100°C, then crushed, ground to fine powder and homogenised by passing through a 1-mm sieve. Samples were sealed in airtight plastic containers of 300 cm^3 and left for more than one month, before counting by gamma ray spectrometry, to allow secular equilibrium between ^{226}Ra and the noble gas radon (^{222}Rn) and its decay products. The parameters altitude and longitude of the locations were obtained based on the Global Positioning System.

2.3 Gamma spectrometry

Each sample was measured with a gamma ray spectrometer consisting of a NaI(Tl) set-up and multichannel analyser 8192 channel, with the following specification: resolution Full Width Maximum Half (FWMH) at 661.6 keV ^{137}Cs (i.e. it is about 40 keV) and about 60 keV for the photopeak of ^{60}Co at 1332 keV. The detector is shielded in a chamber of two layers starting with stainless steel (10-mm thick) and lead (30-mm thick). This shield serves to reduce different background radioactivity.

The spectrometer was calibrated for efficiency and energy using multi-nuclide standard solution (QCY 48) PTB (Germany) (a mixed source containing ^{241}Am, ^{57}Co, ^{60}Co, ^{85}Sr, ^{88}Y, ^{109}Cd, ^{137}Cs, ^{139}Ce and ^{203}Hg)

^{226}Ra activity of the samples was determined via its daughters (^{214}Pb and ^{214}Bi) through the intensity of the 351.93 keV, for ^{214}Pb and 609.31, 1120 and 1764.49 keV, for ^{214}Bi gamma line. ^{232}Th activity of the sample was determined from the daughters (^{228}Ac), (^{212}Pb) and (^{208}Tl) through the intensity of 911.2 keV gamma line for (^{228}Ac), (^{212}Pb) emissions at 238.63 keV and (^{208}Tl) emission at 2614 keV gamma line. ^{40}K activity was determined from the 1460.7 keV emission gamma line. The samples were counted for 12–24 h depending on the concentration of the radionuclides.

The activity concentrations for the natural radionuclides in the measured samples were computed using the following relation (IAEA, 1989).

$$\mathrm{A_{EI}} = NP / t_c \times \mathrm{I}_\gamma\left(\mathrm{E}_\gamma\right) \times \varepsilon\left(\mathrm{E}_\gamma\right) \times \mathrm{M}\left(\mathrm{Bq\,kg^{-1}}\right) \tag{1}$$

where NP is the number of counts in given peak area corrected for background peaks of a peak at energy E, $\varepsilon(E_\gamma)$ is the detection efficiency at energy E, t is the counting lifetime, $I_\gamma(E_\gamma)$ is the number of gamma rays per disintegration of this nuclide at energy E, and M is the mass in kg of the measured sample.

2.4 Calculation of the radiological parameters

To estimate the risk of the radiation hazard of the measured ^{226}Ra, ^{232}Th and ^{40}K activities, many radiological parameters were calculated. The most widely used radiation hazard index is called the radium equivalent activity (Ra_{eq}). The radium equivalent activity is a weighted sum of activities of the ^{226}Ra, ^{232}Th and ^{40}K radionuclides based on the assumption that 370 Bq kg^{-1} of ^{226}Ra, 259 Bq kg^{-1} of ^{232}Th and 4810 Bq kg^{-1} of ^{40}K produce the same gamma ray dose rate (Krisiuk et al., 1971). Radium equivalent activity can be calculated from the following relation suggested by (Beretka and Mathew, 1985).

$$Ra_{eq} = A_{Ra} + (A_{Th} \times 1.43) + (A_K \times 0.077) \quad (2)$$

where A_{Ra} is the activity concentration of ^{226}Ra in Bq kg^{-1}, A_{Th} is the activity concentration of ^{232}Th in Bq kg^{-1} and A_K is the activity concentration of ^{40}K in Bq kg^{-1}.

The second radiation parameter is called the external hazard index (H_{ex}). This criterion considers only the external exposure due to the emitted gamma ray and corresponds to a maximum Ra_{eq} of 370 Bq kg^{-1} for the materials. The value of this index must be less than unity for the radiation hazard to be negligible.

$$H_{ex} = A_{Ra}/370 + A_{Th}/259 + A_K/4810 \leq 1 \quad (3)$$

where A_{Ra}, A_{Th} and A_K are the activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K, respectively. The calculated average external hazard index was found to be less than unity.

In addition to the external hazard, radon and its short-lived products are also hazardous to the respiratory organs. To assess the internal exposure to ^{222}Ra gas and its daughter products, the internal hazard index has been defined by Beretka and Mathew (1985) as:

$$H_{in} = A_{Ra}/185 + A_{Th}/259 + A_K/4810 \leq 1 \quad (4)$$

where A_{Ra}, A_{Th} and A_K are the activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K, respectively. For the safe use of a material in the construction of dwelling, H_{in} should be less than unity.

The total air absorbed dose rate (nGy h^{-1}) due to the mean activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K (Bq kg^{-1}) can be calculated using the formula of Beck et al. (1972) and UNSCEAR (1988):

$$D = 0.462\,A_{Ra} + 0.604\,A_{Th} + 0.042\,A_K \quad (5)$$

where A_{Ra}, A_{Th} and A_K are the mean activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K in Bq kg^{-1}, respectively.

Beck et al. (1972) derived this equation for calculating the absorbed dose rate in air at a height of 1.0 m above the ground from measured radionuclides concentrations in environmental materials.

3 Results and discussion

The distribution of major elements in soils using XRF spectrometry is plotted in Table 1. The result shows homogeneous distribution of the elements in all samples. The only one

exception is sample 55, where it showed the higher composition of Al_2O_3 and Fe_2O_3 (11.14% and 17.37%), respectively; this sample also showed the highest activity concentration of ^{226}Ra compared with other samples. The pH values for the farm surface soil irrigation by flood water from 7.0 to 7.81 were lower than those are for farm surface soil irrigation by groundwater (7.58–8.59). For neutral solutions, the value of pH is 7. If ph > 7, then the solution is alkaline (Tufaila et al., 2006).

Table 1 XRF analysis for some soil samples from Abyan Delta,Yeman, showing their oxides composition

Sample no.	*Type of soil*	*MgO*	*Al_2O_3*	*SiO_2*	*K_2O*	*CaO*	*TiO_2*	*MnO*	*Fe_2O_3*
8	Soil*	5.4823	7.5415	54.6478	2.5037	19.2325	1.9257	0.5443	8.1221
19	Soil*	5.4053	7.3625	54.5122	2.3676	20.2768	2.0474	0.5514	7.4769
29	Soil*	5.3960	7.2678	53.5785	2.3581	21.5485	1.9302	0.5254	7.3956
39	Soil*	8.5644	5.9564	49.9561	2.1209	23.2621	1.9126	0.6755	7.5521
41	Soil*	5.6076	7.1017	52.5909	2.3127	22.1082	1.8509	0.7457	7.6824
46	Soil*	5.8054	5.4823	51.7293	2.5429	21.2527	1.9986	0.5928	8.5612
53	Soil*	5.1610	7.1129	54.4749	2.2925	21.2224	2.0286	0.4946	7.2132
55	Soil*	4.1311	11.1379	42.2034	4.4865	18.1787	1.8693	0.6207	17.3724
2	Soil**	6.4241	7.4400	55.0571	2.4752	18.5071	1.9574	0.5637	7.5753
7	Soil**	5.6630	7.1455	53.4909	2.4315	21.3266	1.8926	0.5443	7.5055
18	Soil**	4.9700	7.1361	55.0853	2.3267	20.3581	2.1221	0.5505	7.4511
28	Soil**	5.1460	7.3652	56.9339	2.4935	18.3094	1.9936	0.5635	7.1950

Note: *farm surface soil irrigation by flood water, **farm surface soil irrigation by groundwater.

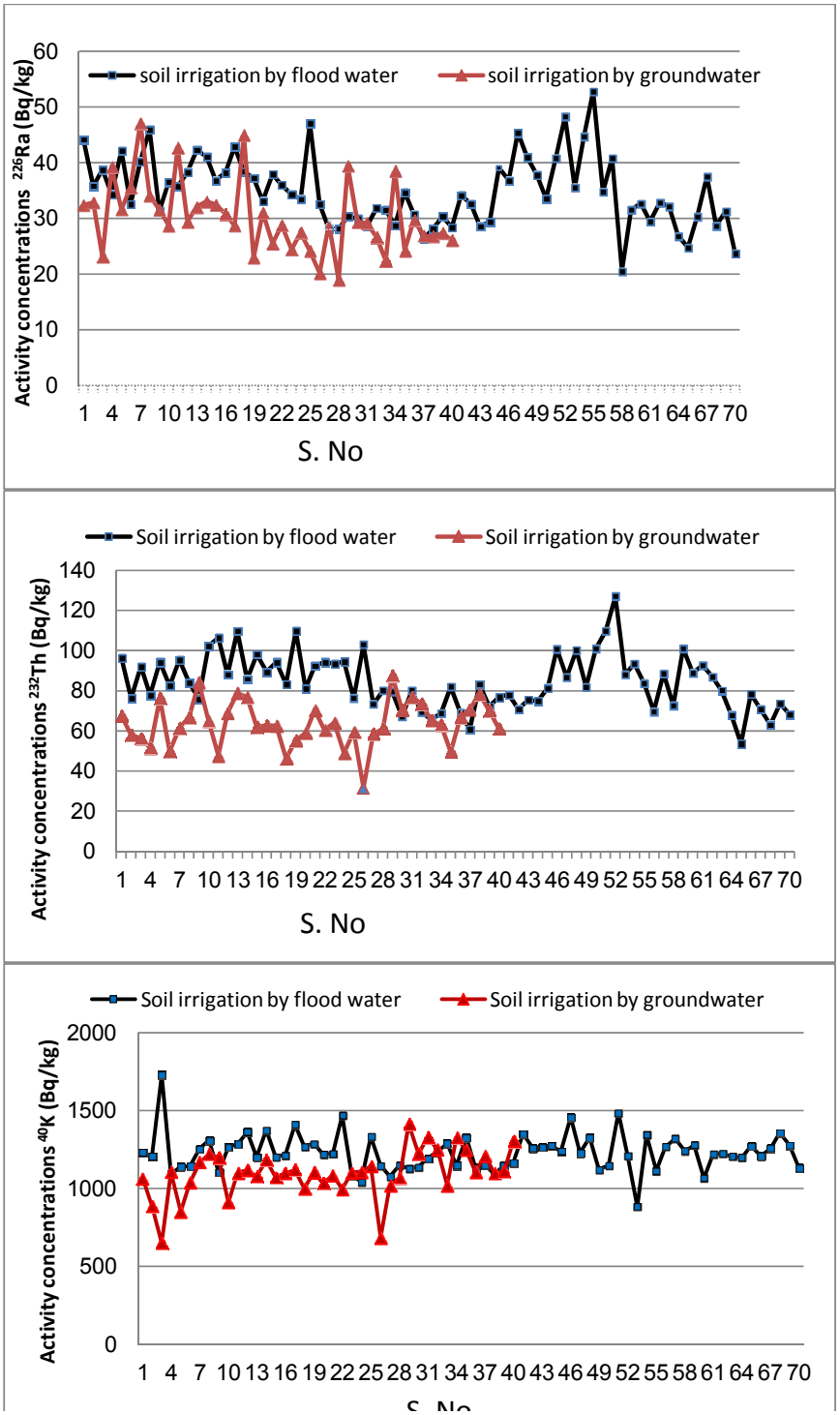
A summary of measurements for the average activity concentration (Bq kg^{-1}) of the natural radioactivity due to ^{226}Ra, ^{232}Th and ^{40}K of farm soil irrigation by flood water and farm soil irrigation by groundwater is given in Table 2. The distribution of ^{226}Ra, ^{232}Th and ^{40}K activity concentrations in all samples is given in Figure 2. It can be concluded that ^{226}Ra ranged from 28.1 ± 3.2 to 52.7 ± 4.3 with an average value of 34.9 ± 2.9 Bq kg^{-1} in farm soils irrigation by flood water samples. The corresponding values are from 19.02 ± 1.5 to 47.11 ± 3.5 with an average value of 30.42 ± 2.7 Bq kg^{-1}, for farm soils irrigation by groundwater samples. ^{232}Th activity concentrations in farm soils irrigation by flood water samples ranged from 53.7 ± 2.2 to 126.7 ± 8.2 with an average value of 84.5 ± 7.8 Bq kg^{-1}. For farm soils irrigation by groundwater the corresponding values are from 31.48 ± 3.7 to 83.93 ± 6.8 with an average value of 63.64 ± 5.7 Bq kg^{-1}. ^{40}K values ranged from 884.2 ± 28.7 to 1486.4 ± 51.7 with an average value of 1232.7 ± 40 Bq kg^{-1} in farm soils irrigation by flood water samples, whereas the corresponding values for farm soil irrigation by groundwater samples are from 685.2 ± 22.2 to 1413.77 ± 49.5 with an average value of 1096 ± 36 Bq kg^{-1}.

Table 2 Activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K (Bq kg^{-1}) of farm surface soil and Urea fertilisers samples and its radiological parameters

Type of soil	^{226}Ra *Min-max* *Mean*	^{232}Th *Min-max* *Mean*	^{40}K *Min-max* *Mean*	Ra_{eq} *(Bq kg^{-1})*	H_{in}	H_{ex}	*Dose rate (nGy h^{-1})*
Soil*	28.1–52.7	53.7–126.7	208.6–322.78	884.2–1486.4	96.8–147.9	0.54–0.87	1.0–0.63
	34.9 ± 2.9	84.5 ± 7.8	250.6 ± 17.1	1232.7 ± 40	118.1 ± 7.9	0.68 ± 0.046	0.77 ± 0.05
Soil**	19.02–47.11	31.48–83.93	117.9–233.8	685.2–1413.77	56.94–129.6	0.42–0.66	0.37–0.84
	30.24 ± 2.7	63.64 ± 5.7	205.7 ± 13.6	1096 ± 36	97.95 ± 6.1	0.56 ± 0.037	0.64 ± 0.04
Urea fertilisers	45.27–33.77	37.1–31.36	–	207.6–391.2	–	–	–
	38.9 ± 4.1	34.3 ±3.4		312.6 ± 13			

Note: *farm surface soil irrigation by flood water, **farm surface soil irrigation by groundwater.

Figure 2 ^{226}Ra, ^{232}Th and ^{40}K activity concentrations for all the soil samples studied (see online version for colours)



Farm soils irrigation by flood water samples shows significantly higher concentrations of ^{226}Ra, ^{232}Th and ^{40}K when compared with farm soils irrigation by groundwater. The flood water helps to washes the radionuclides from the catchment area (10,600 km^2) and deposited them in surface soil of study area (Abyan Delta). The catchment area composed of mid to upper proterozoic metamorphic and intrusive rocks of the southern part of the Arabian Shield. The metamorphic rocks include metavolcanics, amphibole-biotite schists and gneisses, and higher grade migmatites (Kruck et al., 1996). The intrusive rocks include granites and gabbros. Some of these rocks like granite and calc-silicate involve high radioactivity of ^{238}U, ^{232}Th and ^{40}K (Heikal et al., 2007; El-Mageed et al., 2010). Also contamination of flood water may be due to several no nuclear industrial processes: TENORM. These activities can enhance the local activity concentrations such as sewage treatment, gas and oil production and used of coal, oil, and natural gas to produce electric energy (Pöschl and Nollet, 2007).

The variation in concentrations of radionuclides ^{226}Ra, ^{232}Th and ^{40}K for samples under study also regards on regulate of irrigation. Some farm soil received higher amounts of flood water especially those sites in north study area compared with that in south delta. Also the amount of water received by farm soil depends on the type of vegetation, some vegetation need to water more than other, for example banana trees need amounts of water more than lime or guava trees.

Also from the results we noticed the activity concentrations of ^{232}Th were higher than the activity concentrations of ^{226}Ra in all samples under study (^{232}Th content of farm soils irrigation by flood water samples was nearly twice higher when compared with ^{226}Ra content); these results agree with the fact that the concentrations of ^{232}Th radionuclide in earth crust are more abundance (7.4 μg g^{-1}) compared with concentrations of ^{238}U radionuclides (2.8 μg g^{-1}) (UNSCEAR, 2000).

The activity concentration of ^{40}K in soil is higher than that of ^{226}Ra and ^{232}Th for both soils samples, this is also in accordance with the well-known fact that potassium in the earth's crust is of the order of percentage, whereas uranium and thorium are in ppm level (UNSCEAR, 1988). The average concentrations of ^{40}K and ^{232}Th in samples under study were nearly twice higher when compared with the worldwide average concentrations of ^{40}K (412 Bq kg^{-1}) and ^{232}Th (45 Bq kg^{-1}) in soil (UNSCEAR, 2008), while the average activity concentrations of ^{226}Ra for samples under study agree with the worldwide average concentrations of this radionuclide in soil (32 Bq kg^{-1}) (UNSCEAR, 2008).

In this work, urea fertiliser samples (which used in some farm soil especially that irrigation by groundwater) showed that the activity levels were lower than the permissible activity levels which are 32, 45 and 412 Bq kg^{-1} for ^{226}Ra, ^{232}Th, and ^{40}K (UNSCEAR, 2008), respectively (Table 2). Therefore, the use of urea fertiliser does not significant source of radiation hazard and is safe for use in farm soil.

The results for the radium equivalent activity, external hazard index, internal hazard index, and absorbed dose rate of the present work are presented in Table 2. It is observed that the calculated radium equivalent in soils is lower than the allowed maximum value of 370 Bq kg^{-1} (Beretka and Mathew, 1985). The calculated H_{ex} and H_{in} values for the samples under investigation do not exceed the upper limit for H_{ex} and H_{in} which is unity, whereas the absorbed dose rate for soils samples exceed the upper limit for absorbed dose rate which is 59 nG h^{-1}.

To compare the natural radioactivity of Abyan Delta soil with the Yemenian soils in other areas, El-Mageed et al. (2010) study the natural radioactivity of ^{40}K, ^{226}Ra, and ^{232}Th in rock and soil samples collected around Juban town in Yemen; the study shows

the average radioactivity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K for soil samples were 44.4, 58.2, and 822.7 Bq kg^{-1}, respectively. Also El-Kamel et al. (2012) reported that the average specific activities in soil samples collected from Assalamia-Al-Homira area in Abyan governorate were 41.46 ± 5.6, 68.68 ± 6 and 1224.7 ± 31 Bq kg^{-1} for clay and sandy soil, and 80.77 ± 4.5, 211.5 ± 14 and 1004.8 ± 40 Bq kg^{-1} for ^{226}Ra, ^{232}Th and ^{40}K, which reveal that the activity concentrations for sandy soil are much higher than the worldwide average concentrations of this radionuclide in soil of 32, 45 and 412 Bq kg^{-1} for ^{226}Ra, ^{232}Th and ^{40}K, respectively, mentioned by El-Kamel et al. (2012).

Also the activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K in samples from the study area were compared with similar investigations in other countries and the summary of results is given in Table 3. As can be seen from Table 3, ^{232}Th values from the present work for farm surface soil irrigation by flood are higher than that reported by other countries except in case Bangladesh reported by Hamid et al. (2002). In contrast, ^{232}Th values for farm surface soil irrigation by ground water are located in the upper side of the reported values in Table 3. Also ^{226}Ra activity concentrations obtained in this study for both farm surface soil irrigation by flood and farm surface soil irrigation by ground water are lower than values in other countries except in cases Egypt (Qena) reported by Ahmed and El-Arabi (2005) and Saudi Arabia (Taif) reported by El-Aydarous (2007). On the other hand, ^{40}K values in the present study are higher than values in other countries except in cases Bangladesh reported by Hamid et al. (2002) and Malaysia reported by Saleh et al. (2013).

Table 3 Comparison of mean activity concentrations (Bq kg^{-1}) in soil with other countries of the world

Country	*^{226}Ra (Bq kg^{-1})*	*^{232}Th (Bq kg^{-1})*	*^{40}K (Bq kg^{-1})*	*References*
Bangladesh	84	141	1944	Hamid et al. (2002)
Egypt (Qena)	13.7	12.3	162.8	Ahmed and El-Arabi (2005)
India	44.97	59.7	217	Dhawal et al. (2013
Jordan (Northern Jordan)	42.5	26.7	291.1	Al-Hamarneh et al. (2009)
Jordan (Ma'an)	57.7	18.1	138.1	Saleh and Shayeb (2014)
Lebanon	4–73	5–50	57–554	El Samad et al. (2013)
Malaysia	12-968	11-1210	12-2450	Saleh et al. (2013)
Saudi Arabia (Taif)	23.8	18.6	162.8	El-Aydarous (2007)
Yemen (Juban)	44.4	58.2	822.7	El-Mageed et al. (2010)
Yemen (Assalamia-Al-Homira) clay	41.46	68.68	1224.7	El-Kamel et al. (2012)
Farm soil*	34.9	84.5	1232.7	Present study
Farm soil**	30.24	63.64	1096	
Worldwide	32	45	412	UNSCEAR (2008)

Note: *farm surface soil irrigation by flood water, **farm surface soil irrigation by groundwater.

4 Conclusion

1 Surface soil of the Abyan Delta in Yemen was measured for its radioactivity content. The results show that the mean concentration values of ^{226}Ra, ^{232}Th and ^{40}K in farm surface soil irrigation by flood water were 34.9, 84.5 and 1232.7 Bq kg^{-1}, while that of farm surface soil irrigation by groundwater samples were 30.24, 63.64, 1096 Bq kg^{-1}, respectively.

2 The means of radium equivalent activity (Ra_{eq}), external hazard index and internal hazard index for all samples under investigation – farm surface soil irrigation by flood water and farm surface soil irrigation by groundwater – are 250.6, and 205.7 Bq kg^{-1} for Ra_{eq} and 0.68, 0.56 for H_{ex} and 0.77, 0.64 for H_{in}, respectively.

3 The results indicate that the dose rates at 1 m above the ground from terrestrial sources in all samples under investigation were 118.1 and 97.95 nGy h^{-1} for farm surface soil irrigation by flood water and farm surface soil irrigation by groundwater, respectively. These values are higher than the estimate of average global terrestrial radiation of 55 nGy h^{-1}.

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