

# Radioactivity of <sup>238</sup>U, <sup>232</sup>Th, <sup>40</sup>K, and <sup>137</sup>Cs and assessment of depleted uranium in soil of the Musandam Peninsula, Sultanate of Oman

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#### Abstract

<sup>238</sup> U, <sup>232</sup> Th, <sup>40</sup> K, and <sup>137</sup> Cs radioactivities were determined in soil samples collected from the Musandam Peninsula, Sultanate of Oman, using a high resolution gamma-ray spectroscopy system. In addition, total uranium was measured in selected soil samples by inductively coupled plasma optical emission spectroscopy (ICP-OES). The averages of radioactivity levels in the soil for <sup>238</sup> U, <sup>232</sup> Th, and <sup>40</sup> K were 14.42 Bq kg<sup>-1</sup>, 9.95 Bq kg<sup>-1</sup>, 158.21 Bq kg<sup>-1</sup>, and 2770 Bq m<sup>-2</sup> for <sup>137</sup> Cs, respectively. The mean of the total uranium in soils was 1578.28  $\mu$ g kg<sup>-1</sup>. As the mean values, radium equivalent was estimated to be 40.84 Bq kg<sup>-1</sup> and annual effective outdoor radiation dose was 25.4  $\mu$ Sv. The contributions of individual radionuclides for <sup>238</sup> U, <sup>232</sup> Th, <sup>40</sup> K, and <sup>137</sup> Cs to the total annual effective dose were 30%, 32%, 32%, and 6% respectively. Selected soil samples were also investigated for depleted uranium (DU) by estimating the percentages of <sup>235</sup> U in total uranium and the results revealed that DU concentrations matched its range in natural uranium.

Key Words: Soil, Oman, uranium, cesium-137, radium-226, thorium-232, potassium-40

## 1. Introduction

Radiation and radioactivity in the environment have natural and man-made sources. Exposure to natural radiation represents the most significant part of the total exposure to radiation in the environment (Tso and Leung, 2000; UNSCEAR, 2008). Natural radionuclides in soil generate a significant component of the background radiation that the population is exposed to (Goddard, 2002). Only natural radionuclides with half-lives comparable with the age of the Earth or their corresponding decay products existing in terrestrial material such as <sup>232</sup>Th, <sup>238</sup>U, <sup>235</sup>U, and <sup>40</sup>K are of great interest. The levels of these radionuclides are relatively distributed in soil based on the nature of its geological formations (Al-Jundia et al., 2003; Orabi et al., 2006).

Human activities like mining and milling of mineral ores, processing and enrichment, nuclear fuel fabrications, and handling of the fuel cycle tail end products cause release of additional amounts of natural radionuclides into the environment (UNSCAR, 1993). Human beings are exposed outdoors to the natural terrestrial radiation

that originates predominantly from the upper 30 cm of the soil only (Chikasawa et al., 2001). Knowledge of their distribution in soil and rock plays an important role in the field of radiation protection (Rani and Singh, 2005).

Man-made radionuclides are produced from nuclear industrial activities, nuclear power plant accidents, or military uses. Radionuclides produced from nuclear activities are dispersed into the environmental ecosystems depending on the physicochemical characteristics of the radionuclides and the environmental prevailing conditions (UNSCAR, 1993).

 $^{137}$ Cs (T<sub>1/2</sub> = 30.17 years) is a man-made radionuclide released from nuclear fission and activation processes. The latitude and rate of precipitation are the main factors affecting the distribution of  $^{137}$ Cs on the Earth's surface (Ritchie and McHenry, 1990). The high fission yields as well as longer half-life allow a good portion of the originally released  $^{137}$ Cs to be with us today. A large amount of this radionuclide was released into the atmosphere during the nuclear weapons tests in the 1950s and 1960s with a minor period of deposition from 1971 to 1974 (Ritchie and McHenry, 1990). The atmospheric deposition of  $^{137}$ Cs from above-ground nuclear weapons testing has made it a typical background component of northern hemisphere topsoil samples (Ebert et al., 1990). Deposition of  $^{137}$ Cs released during nuclear reactor accidents is more localized and strongly depends on meteorological conditions (Al-Mari, 2006). Evidence of possible transfer and accumulation of the  $^{137}$ Cs radionuclide in the soil of Burullus Lake in Egypt was presented by El-Reefy et al. (2006). Vertical distribution and inventories of  $^{137}$ Cs have been determined using radiocesium distributions in presumably undistributed soil profiles collected from 36 sites distributed all over Syria (eastern Mediterranean region) (Al-Marsi, 2006).

Depleted uranium (DU) is a waste product of uranium enrichment and is distinguished from natural uranium by lower relative concentrations of  $^{235}$ U (<0.7%) and  $^{234}$ U. DU has civilian and military applications. The typical civilian application is in the area of radiation protection as a shield because of its high density (19.07 g/cm<sup>3</sup>) and high atomic number (z = 92). It is also used as a counterweight and ballast in aircraft and yachts and as a catalyzer in chemical processes. The military applications of DU are in ammunition and as an element of guided missiles (Nada et al., 2001).

Determination and assessment of radioactivity in the environment has benefits for the following:

(1) Establishing baseline data on the level of radionuclide, (2) Serving as the reference in documenting changes to environmental radioactivity in soil due to anthropogenic activities, (3) Estimation of the radiation dose distribution is important in assessing the health risk to a population (Obed et al., 2005).

The aim of this paper is to present the first stage (radioactivity levels in soil) of a comprehensive program, whose purpose is the determination and assessment of the radioactivity levels in the Sultanate of Oman environment. This program includes soil, beach sediments, vegetation covers, and underground and surface waters samples. Selection of the Musandam Peninsula region in this work was due to its vulnerable location on the west coast of the Strait of Hormuz at the entrance to the Arabian/Persian Gulf. In addition, the region is near Bushehr (the location of Iran's nuclear power site) and is separated by the Gulf of Oman and the Arabian Sea from Pakistan's and India's nuclear sites. In addition, the importance of this radiological study is to evaluate the effect of the Gulf War (1990/1991) on the Musandam region because about 321 t of DU had been used by the US forces during the war (Bleise et al., 2003). This amount of DU is much greater than that used in Bosnia-Herzegovina (approximately 3 t) and also in Kosovo (>10 t) (DoAF, 1997; Harley et al., 1999; Hamilton, 2000).

## 2. Sites description and sampling locations

The Musandam Peninsula, covering  $1800 \text{ km}^2$ , is in the far north of the Sultanate of Oman, embossed into the Strait of Hormuz at the entrance to the Arabian/Persian Gulf (Figure 1). The Musandam Peninsula takes the shape of an acute-angled isosceles triangle; its head lies on the strategic Strait of Hormuz. The nearest point to Iran across the strait is not more than 55 km and does not exceed 60 m in depth. The peninsula has 2 coasts: western coast on the Arabian/Persian Gulf and eastern coast on the Gulf of Oman, connected to the Indian Ocean (Rodrigue, 2004). The strait has economic and political importance: about 40% of the world's seaborne oil shipments and 20% of all world shipments and about 15 tankers carrying 16.5 to 17 million barrels of crude oil normally pass through the strait every day (U.S. Energy Information Administration, 2008). The land around the main city of Al-Khasab is used for agriculture. The main industrial activities are fishing and ship building. Apart from Al Khasab there are small settlements and the other areas are barren mountains.



Figure 1. Map of the Musandam Peninsula and the soil sampling locations.

#### 3. Sample preparation and measurement

Thirty-three sampling locations were chosen for soil sampling during the period of 2007-2010 (Figure 1). Nine soil samples were collected from each location of about  $120 \text{ m}^2$  and were taken from the surface layer soil of 10 cm depth. The studied locations include urban areas, valleys, and mountains. The samples include alluvial loam from valleys, sands, and bulk soil from undisturbed, uncultivated, and grass covered areas. The collected samples were sent to the radiation measurement laboratory of the Ministry of Regional Municipalities and Water Resources. The 9 samples from each location were cleaned and mixed thoroughly to form 1 composite sample and then homogenized using a mechanical mixer/divider. Each homogenized sample was dried at room

temperature for a few days and then in an oven at 105 °C for about 3 days. One representative sample was taken from each composite to represent the location using a mechanical divider. The representative samples were grinded with a grinder to pass through a 100-mm sieve. In order to maintain radioactive equilibrium between  $^{226}$ Ra and its daughters the soil samples were sealed in 500-mL Marinelli beakers and then weighed and stored for 1 month. Gamma-ray spectrometric analysis was done using an extended range reverse electrode closed end germanium detector with a relative efficiency of 60%. The resolutions expressed in FWHMs of photopeaks are 0.79 keV at the 122 KeV  $\gamma$ -peak of <sup>57</sup>Co and 1.89 keV at the 1332 keV  $\gamma$ -peak of <sup>60</sup>Co, respectively. The detector was housed inside a 10 cm lead shield to reduce the background radiation and was concentric with a thin layer of copper. Data acquisition was performed by using an 8 K multichannel analyzer and the data were analyzed by computer software (Genie-2000).

The energy calibration was done by using certified standards (<sup>241</sup> Am, <sup>57</sup> Co, <sup>137</sup> Cs, and <sup>60</sup> Co). The efficiency calibrations were done by using certified multi-gamma rays soil standard containing <sup>238</sup> U, <sup>241</sup> Am, <sup>134</sup> Cs, <sup>137</sup> Cs, <sup>65</sup> Zn, <sup>54</sup> Mn, and <sup>40</sup> K packed in a 500-mL Marinelli beaker. All certified standards were obtained from Canberra Industries Company. Gamma ray spectrum of a blank sample (double distilled deionized water of >15 M $\Omega$ ) was subtracted from each sample's gamma spectrum.

The activity of <sup>238</sup>U (<sup>226</sup>Ra) was determined from the 609.3, 1120.3, and 1764.5 keV gamma-ray peaks of <sup>214</sup>Bi and from the 295.2 and 351.9 keV gamma-ray peaks of <sup>214</sup>Pb. On the other hand, the 911.2, 964.6, and 969.0 keV gamma-ray peaks of <sup>228</sup>Ac and the 583.2 keV of <sup>208</sup>Tl were used to evaluate <sup>232</sup>Th. <sup>40</sup>K and <sup>137</sup>Cs were determined using 1460.8 and 661.7 keV, respectively. The activity of <sup>235</sup>U was determined in 16 dried samples that contained high levels of <sup>238</sup>U to obtain reasonable intensities for the 143.76 keV gamma-ray peak. In addition, the 185.72 keV gamma-ray peak was used after interference correction according to Papachristodoulou et al. (2003). The minimum detectable activities (MDAs) of measured radionuclide were calculated based on the counting method parameters and are given in Table 1.

Radionuclide	E (keV)	MDA (Bq $kg^{-1}$ )
$^{238}$ U ( $^{226}$ Ra)	295.2	0.22
	351.9	0.16
	609.3	0.08
	1121.3	0.47
	1764.5	0.25
$^{232}$ Th	583.2	0.07
	911.2	0.26
	964.6	0.88
$^{40}\mathrm{K}$	1460.8	0.57
$^{137}Cs$	661.7	0.05
$^{235}\mathrm{U}$	143.76	0.62
	185.6	0.15

Table 1. The minimum detectable activities (MDA) corresponding to the gamma rays of radionuclides with 28,800 s counting time and 500-mL Marinelli beaker.

For determination of total U, 10 g from each sample used for  $^{235}$ U determination was placed in a high temperature furnace (550 °C) overnight to remove the organic components. Three replicate aliquots of soil residue (0.4 g of each) were taken from each sample and then digested with HNO<sub>3</sub>/HF/HClO<sub>4</sub>, followed by dilution and filtration, and then measured by inductively coupled plasma optical emission spectroscopy (ICP-

OES). The calibration of ICP-OES was done by using a set of uranyl acetate concentrations covering the range of interest (0.1-100 mg). The laboratory measurements were subjected to a regular quality control program including periodical monitoring of the backgrounds, calibrations, and the detection system performances. Quality control soil samples MaS16, MaS17, and MaS18 were regularly measured in parallel with the soil samples. These quality control samples were received by the laboratory through the participations in the Mixed Analyte Performance Program (MAPEP), organized by the US Department of Energy (US DOE).

## 4. Results and discussion

The activity concentrations of the detected <sup>238</sup>U, <sup>232</sup>Th, <sup>40</sup>K, and <sup>137</sup>Cs in parallel with the ground position coordinates (longitudes and latitudes) for the studied locations are presented in Table 2. It was observed that  $^{238}$ U activity in soil was higher than that of  $^{232}$ Th and ranged between 5.45 Bq kg<sup>-1</sup> and 25.7 Bq kg<sup>-1</sup> with an average of 14.42 Bq kg<sup>-1</sup>. In contrast, <sup>232</sup>Th activity was lower than that of <sup>238</sup>U and ranged between 0.88 Bq kg<sup>-1</sup> and 20.18 Bq kg<sup>-1</sup> with an average of 9.95 Bq kg<sup>-1</sup>. The activity of  ${}^{40}$ K was higher than that of both  $^{238}$ U and  $^{232}$ Th in all studied locations and its levels ranged from 10.49 Bq kg<sup>-1</sup> to 282.86 Bq kg<sup>-1</sup> with an average of 158.21 Bq kg<sup>-1</sup>. Figures 2-4 show the frequency distributions of <sup>238</sup>U, <sup>232</sup>Th, and <sup>40</sup>K, respectively. It is clear that both  $^{238}$ U and  $^{232}$ Th distributions have a slight positive skew (0.09) while a negative skew (-0.5) is shown for  $^{40}$ K. The levels of detected radionuclides in soil samples indicated wide variations and this may be attributed to the diversity of formations and textures of the soil in the studied area. The soil in most of studied valleys is layer plates of clay that resulted from runoff of rain water while other samples from mountains were mainly sands of different grain sizes and colors. A strong linear correlation coefficient of 0.84was detected between <sup>238</sup>U and <sup>232</sup>Th levels, of 0.82 between <sup>40</sup>K and <sup>238</sup>U, and of 0.84 between <sup>40</sup>K and  $^{232}$ Th. This may be attributed to the limitation of activities that alter the natural compositions of soil such as agricultural or industrial activities in the region. It is well known that agricultural activities are frequently associated with fertilization of soil with phosphate that contains elevated levels of U, Th, and K. The industrial activities such as mining processes concentrate and redistribute U and Th in the environment. However, the variability among levels of <sup>238</sup>U and levels <sup>232</sup>Th are frequently associated with the type of geological minerals. Therefore, detailed mineralogical investigations are needed for more interpretations.





Figure 2. The specific activity levels distribution of  $^{238}$  U in the investigated soil samples. Skewness = 0.09 and kurtosis = -0.07.

Figure 3. The specific activity levels distribution of  $^{232}$  Th in the investigated soil samples. Skewness = 0.09 and kurtosis = -0.11.

Location	Lat	Long	Activity concentration dry weight				
Location	Lat.	Long.	$^{238}U$	$^{232}$ Th	<sup>40</sup> K	$^{137}Cs$	
L1	$26^{\circ}03'26''$	$56^{\circ}05'12''$	$25.70 \pm 1.5$	$19.80\pm0.7$	$262.81 \pm 3.20$	$377.5 \pm 26.5$	
L2	$26^{\circ}04'34''$	56°05′52″	$12.19\pm0.8$	$8.11\pm0.46$	$161.11 \pm 2.04$	$1107.5\pm35$	
L3	26°06′26″	56°07′26″	$5.45 \pm 0.25$	$1.25\pm0.08$	$17.10 \pm 0.50$	$280 \pm 22.5$	
L4	$26^{\circ}06'05''$	56°07′27″	$17.05 \pm 0.85$	$7.09\pm0.3$	$150.21 \pm 2.05$	$377.5 \pm 25.5$	
L5	$56^{\circ}07'43''$	26°07′28″	$11.71\pm0.9$	$7.01\pm0.31$	$130.68 \pm 2.72$	$1645 \pm 35$	
L6	$56^{\circ}08'25''$	26°07'31"	$6.65\pm0.33$	$1.49 \pm 0.09$	$56.60 \pm 1.47$	$192.5 \pm 10$	
L7	56°08′50″	$26^{\circ}07'44''$	$12.07 \pm 0.73$	$5.52\pm0.21$	$166.60 \pm 2.30$	$27.5\pm5$	
L8	$56^{\circ}10'38''$	$26^{\circ}10'58''$	$16.34 \pm 0.84$	$11.26 \pm 0.41$	$180.31 \pm 3.47$	$2955\pm30$	
L9	56°10′59″	$26^{\circ}13'10''$	$17.73\pm0.81$	$10.96 \pm 0.40$	$210.25 \pm 3.49$	$5080 \pm 120$	
L10	$56^{\circ}13'18''$	26°11′46″	$21.28 \pm 1.2$	$13.90 \pm 0.54$	$213.71 \pm 3.97$	$5562.5\pm80$	
L11	$56^{\circ}14'53''$	26°11′46″	$15.14 \pm 0.82$	$9.01\pm0.47$	$154.92 \pm 2.01$	$840 \pm 115$	
L12	56°15′00″	26°10'26"	$18.92\pm0.8$	$11.60 \pm 0.41$	$201.51 \pm 2.55$	$2305\pm20$	
L13	$56^{\circ}15'11''$	26°11′57″	$11.88 \pm 0.94$	$7.44 \pm 0.32$	$133.6\pm2.5$	$1927.5\pm37$	
L14	56°16′51″	26°13'00"	$18.84\pm0.81$	$11.13 \pm 0.49$	$185.12 \pm 3.35$	$15350 \pm 52.5$	
L15	$56^{\circ}16'92''$	26°12'93"	$15.73 \pm 0.84$	$10.80 \pm 0.44$	$175.60 \pm 3.1$	$5270\pm55$	
L16	$56^{\circ}19'44''$	$26^{\circ}16'40''$	$9.51\pm0.57$	$3.46\pm0.15$	$59.20\pm0.86$	$3052.5\pm27$	
L17	$56^{\circ}25'51''$	26°20′47″	$9.45\pm0.55$	$12.06 \pm 0.4$	$210.29 \pm 3.59$	$1565 \pm 15$	
L18	$56^{\circ}25'53''$	$26^{\circ}20'44''$	$15.21\pm0.8$	$13.60 \pm 0.51$	$231.79 \pm 2.66$	$1765 \pm 17.5$	
L19	56°31′40″	26°22′57″	$19.01\pm0.81$	$17.51\pm0.8$	$282.86 \pm 3.50$	$9225 \pm 47.5$	
L20	$56^{\circ}19'11''$	$25^{\circ}45'45''$	$9.13 \pm 0.5$	$5.75\pm0.2$	$69.47 \pm 1.30$	$82.5 \pm 5.5$	
L21	56°19′58″	26°09'44"	$7.83 \pm 0.34$	$0.88\pm0.05$	$10.49 \pm 0.33$	$87.5\pm5$	
L22	56°19′57″	26°09'46"	$14.44 \pm 0.73$	$11.25\pm0.32$	$185.72 \pm 3.14$	$3260 \pm 37.5$	
L23	$56^{\circ}19'97''$	26°05'77"	$17.14 \pm 0.82$	$20.18 \pm 0.7$	$123.88 \pm 3.74$	$137.5 \pm 10$	
L24	$56^{\circ}21'62''$	$26^{\circ}02'34''$	$11.61\pm0.95$	$7.64 \pm 0.33$	$125.21 \pm 2.06$	$2587.5 \pm 251$	
L25	$56^{\circ}21'97''$	26°02'69"	$18.07\pm0.85$	$14.35 \pm 0.46$	$225.14 \pm 4.21$	$7052.5 \pm 77.5$	
L26	$56^{\circ}21'84''$	$26^{\circ}02'71''$	$13.32 \pm 0.72$	$10.02 \pm 0.41$	$151.63 \pm 3.4$	$1587.5 \pm 37.5$	
L27	56°19′54″	26°04′48″	$19.61 \pm 0.79$	$13.72 \pm 0.5$	$200.40 \pm 3.77$	$4767.5 \pm 75$	
L28	$56^{\circ}25'88''$	$25^{\circ}56'16''$	$11.94\pm0.9$	$8.34\pm0.44$	$114.21 \pm 1.89$	$2870 \pm 12.5$	
L29	56°25′83″	$25^{\circ}54'52''$	$13.66 \pm 0.7$	$7.17\pm0.18$	$130.99 \pm 2.19$	$3380 \pm 45.5$	
L30	$56^{\circ}19'11''$	$25^{\circ}45'45''$	$9.13\pm0.49$	$5.75\pm0.21$	$69.47 \pm 1.47$	$135\pm5.5$	
L31	56°15′86″	25°39'23"	$16.64\pm0.8$	$15.44 \pm 0.4$	$237.64 \pm 4.27$	$1632.5 \pm 40$	
L32	$56^{\circ}12'30''$	$25^{\circ}57'40''$	$15.54 \pm 0.79$	$11.62\pm0.33$	$178.14 \pm 3.20$	$3190 \pm 45.5$	
L33	56°12'30"	25°57′40″	$17.96 \pm 0.8$	$13.37\pm0.53$	$214.12 \pm 3.62$	$1767.5\pm35$	
	Ave.		14.42	9.95	158.21	2770	
	Max.		25.7	20.18	282.86	$15,\!350$	
	Min.		5.45	0.88 10.49		27.5	
	SD.		4.52	4.84	67.26	3150	
	Skewness		0.089	0.089	-0.496	2.362	
	Kurtosis		-0.067	-0.112	-0.171	7.299	

**Table 2.** Geographical locations and activity concentrations of  $^{238}$  U,  $^{232}$  Th, and  $^{40}$  K in Bq kg<sup>-1</sup> and  $^{137}$  Cs in Bq m<sup>-2</sup> in the studied soil samples.

Comparing internationally, the measured levels of  $^{238}$  U,  $^{232}$  Th, and  $^{40}$  K are lower than the world averages of 40 Bq kg<sup>-1</sup> for  $^{238}$  U, 40 Bq kg<sup>-1</sup> for  $^{232}$  Th, and 370 Bq kg<sup>-1</sup> for  $^{40}$  K (UNSCEAR, 2000). The calculation indicated that the levels in the present study were 0.36, 0.25, and 0.43 times of the world averages for  $^{238}$  U,  $^{232}$  Th, and  $^{40}$  K, respectively.

Table 4 compares the reported values of  $^{238}$ U,  $^{232}$ Th, and  $^{40}$ K in the soil in different countries (UN-SCEAR, 2000) with those determined in the present study. It is found that the ranges of  $^{238}$ U and  $^{232}$ Th almost match those of Egypt, Belgium, Poland, and China and are in the lower range compared with those of the other countries. However, the values of  $^{40}$ K are in the lower range when compared worldwide.

 $^{137}$ Cs was detected in all sites with significant variations. Its levels ranged from 27.5 Bq m<sup>-2</sup> to 15,350 Bq m<sup>-2</sup> with the average of 2770 Bq m<sup>-2</sup>. The frequency distribution of  $^{137}$ Cs activities in tested samples indicated a positive skew of 2.36 as shown in Figure 5. The variability in its levels is frequently associated with the soil texture. The high levels were observed in undisturbed and grass-covered areas. This may be attributed to the clay soil, which contains residues of biogenic organic matter and has an ability to retain  $^{137}$ Cs nuclide. The strong retention of  $^{137}$ Cs at the surface of different soils frequently is due to the presences of clay minerals and organic matter (Price, 1991; Holgye and Maly, 2000; Arapis and Karandinos, 2004; Sigurgeirsson et al., 2005). Organic matter contents were suggested to affect the retention and migration of the fallout radionuclides in the environment (e.g., De Brouwer et al., 1994; Staunton et al., 2002; El-Reefy et al., 2006). A hypothesis was also suggested that organic matter modifies the adsorption properties of clay minerals in soil (e.g., Staunton et al., 2002).





Figure 4. The specific activity levels distribution of  ${}^{40}$  K in the investigated soil samples. Skewness = -0.5 and kurtosis = -0.17.



# 4.1. Calculation of radiological effects

Radiological effects of external radiation exposure can be assessed for the naturally occurring radioactive materials (NORM)  $^{238}$  U,  $^{232}$  Th, and  $^{40}$  K by deducing the radium equivalent (Ra<sub>eq</sub>). Calculating the absorbed dose rates (D) and the annual effective dose (H) is also useful for calculating the contribution the NORM with other sources of background radiation. External radiation hazard index (H<sub>ex</sub>) is considered an index for the significance of radiation exposures.

# 4.1.1. Radium equivalent $(Ra_{eq})$

Since the diversity levels of NORM in the soil samples are not uniform, it will be convenient to use the term  $\operatorname{Ra}_{eq}$  in Bq kg<sup>-1</sup> to compare the specific activity of materials containing different amounts of <sup>238</sup>Ra, <sup>232</sup>Th, and <sup>40</sup>K. The (Ra<sub>eq</sub>) activities were calculated according to Eq. (1) (Beretka and Mathew, 1985; Hayambu et al., 1995):

$$Ra_{eq} = C_{Ra} + 1.43C_{Th} + 0.077C_K,\tag{1}$$

where  $C_{Ra}$  is the specific activity of  $^{226}$ Ra ( $^{238}$ U) in Bq kg<sup>-1</sup>,  $C_{Th}$  is the specific activity of  $^{232}$ Th in Bq kg<sup>-1</sup>, and  $C_K$  is the specific activity of  $^{40}$ K in Bq kg<sup>-1</sup>.

Table 3. Radium equivalent (Ra  $_{eq}$ ) in Bq kg  $^{-1}$ , the absorbed dose rates (D) in nGy h  $^{-1}$ , annual effective dose (H) in  $\mu$ Sv y  $^{-1}$  for  $^{238}$  U,  $^{232}$  Th,  $^{40}$  K, and  $^{137}$  Cs and the external radiation hazard index (H  $_{ex}$ ) for all investigated locations.Location $\frac{238}{Ra_{eq}}$  D H H $_{ex}$  D HTotal HL174.2534.7942.700.200.170.2142.91

Location	200	0, 2021	n and $10$	TOTUS		Total H	
Location	$\operatorname{Ra}_{eq}$	D	Η	$H_{ex}$	D	Η	1004111
L1	74.25	34.79	42.70	0.20	0.17	0.21	42.91
L2	36.19	17.25	21.17	0.10	0.50	0.61	21.78
L3	8.55	3.99	4.89	0.02	0.13	0.15	5.05
L4	38.75	18.42	22.61	0.10	0.17	0.21	22.82
L5	31.80	15.09	18.52	0.09	0.74	0.91	19.43
L6	13.14	6.33	7.77	0.04	0.09	0.11	7.88
L7	32.79	15.86	19.46	0.09	0.01	0.02	19.48
L8	46.33	21.87	26.84	0.13	1.33	1.63	28.47
L9	49.59	23.58	28.94	0.13	2.29	2.81	31.74
L10	57.61	27.14	33.31	0.16	2.50	3.07	36.38
L11	39.95	18.90	23.19	0.11	0.38	0.46	23.65
L12	51.02	24.15	29.64	0.14	1.04	1.27	30.91
L13	32.81	15.55	19.09	0.09	0.87	1.06	20.15
L14	49.01	23.15	28.41	0.13	6.91	8.48	36.88
L15	44.70	21.11	25.91	0.12	2.37	2.91	28.82
L16	19.02	8.95	10.99	0.05	1.37	1.69	12.67
L17	42.89	20.42	25.06	0.12	0.70	0.86	25.92
L18	52.51	24.91	30.57	0.14	0.79	0.97	31.54
L19	65.83	31.15	38.23	0.18	4.15	5.09	43.33
L20	22.70	10.59	12.99	0.06	0.04	0.05	13.04
L21	9.90	4.59	5.63	0.03	0.04	0.05	5.68
L22	44.83	21.21	26.03	0.12	1.47	1.80	27.83
L23	55.54	25.27	31.02	0.15	0.06	0.08	31.09
L24	32.18	15.20	18.65	0.09	1.16	1.43	20.08
L25	55.93	26.40	32.40	0.15	3.17	3.89	36.30
L26	39.32	18.53	22.74	0.11	0.71	0.88	23.62
L27	54.66	25.70	31.54	0.15	2.15	2.63	34.18
L28	32.66	15.32	18.80	0.09	1.29	1.58	20.38
L29	34.00	16.10	19.76	0.09	1.52	1.87	21.63
L30	22.70	10.59	12.99	0.06	0.06	0.07	13.07
L31	57.02	26.92	33.04	0.15	0.73	0.90	33.94
L32	45.87	21.63	26.54	0.12	1.44	1.76	28.30
L33	53.57	25.30	31.05	0.14	0.80	0.98	32.03
Ave	40.84	19.27	23.87	0.11	1.25	1.53	25.4
Max.	74.25	34.79	42.70	0.20	6.91	8.48	43.33
Min.	8.55	3.99	4.89	0.02	0.01	0.02	5.05
SD.	15.70	7.38	9.05	0.04	1.42	1.74	9.90

This equation is based on the fact that 370 Bq kg<sup>-1</sup> of <sup>226</sup>Ra or 259 Bq kg<sup>-1</sup> of <sup>232</sup>Th or 4810 Bq kg<sup>-1</sup> of <sup>40</sup>K produce the same gamma dose rate and assuming radioactive equilibrium to be established in both <sup>238</sup>U series and <sup>232</sup>Th series (Yu et al., 1988). The obtained values of Ra<sub>eq</sub> (Table 3) ranged from 8.55 Bq kg<sup>-1</sup> to 74.25 Bq kg<sup>-1</sup> with an average of 40.84 Bq kg<sup>-1</sup>. The minimum and maximum values are due to the soil from L3 and L1, respectively. All soil samples recorded radium equivalent levels far below the limit (370 Bq kg<sup>-1</sup>) for exposure to the NORM (OECD, 1979). Therefore, the soil from this region is safe and can be used as construction materials without posing any significant radiological threat to the population.

Pagion /	Concentration in soil (Bq $kg^{-1}$ )						Absorbed dose rates	
Country	<sup>226</sup> Ra		$^{232}$ Th		$^{40}\mathrm{K}$		in air (nGy $h^{-1}$ )	
Country	Mean	Range	Mean	Range	Mean	Range	Mean	Range
Egypt	17	5-64	18	2-96	320	29-650	32	20-133
United States	40	8-160	35	4-130	370	100-700	47	14-118
India	29	7-81	64	14-160	400	38-760	56	20-1100
Japan	33	6-98	28	2-88	310	15-990	53	35-70
China	32	2-440	41	1-360	440	9-1800	62	2-340
Luxembourg	35	6-52	50	7-70	620	80-1800	49	14-73
Iran	28	8-55	22	5-42	640	250-980	71	36-130
Denmark	17	9-29	19	8-30	460	240-610	52	35-70
Belgium	26	5-50	27	5-50	380	70-900	43	13-80
Greece	25	1-240	21	1-190	360	12-1570	56	30-109
Spain	32	6-250	33	2-210	470	25-1650	76	40-120
Switzerland	40	10-900	25	4-70	370	40-1000	45	15-120
Portugal	44	8-65	51	22-100	470	25-1650	76	40-120
Bulgaria	45	12-210	30	7-160	400	40-800	70	48-96
Romania	32	8-60	38	11-75	490	250-1100	59	21-122
Poland	26	5-120	21	4-77	410	110-970	45	18-97
Present study	14	5-26	10	1-20	158	10-283	19	4-35

**Table 4.** Comparison of natural radioactivity levels in soil and air-absorbed dose in Musandam (Oman) with those in other countries given in UNSCEAR (2000).

# 4.1.2. External radiation hazard index $(H_{ex})$ of NORM

The external radiation hazard index  $(H_{ex})$ , also called the representative level index, is defined from Eq. (2) (Beretka and Mathew, 1985):

$$H_{ex} = Ra/370 + C_{Th}/259 + C_K/4810 \le 1,$$
(2)

where  $C_{Ra}$ ,  $C_{Th}$  and  $C_K$  have the same meaning as in Eq. (1).

The values of this index are listed in Table 3. The levels range from 0.02 to 0.2 with an average of 0.11. The minimum and maximum are recorded for L3 and L1, respectively. However, the value of this index must be less than unity in order to keep the radiation hazard insignificant.

# 4.1.3. The dose rates from NORM

The total air absorbed dose rate in nGy  $h^{-1}$  in air at 1 m above the ground surface due to the activity concentrations of <sup>238</sup>U, <sup>232</sup>Th and <sup>40</sup>K (Bq kg<sup>-1</sup>) can be calculated using Eq. (3) (UNSCEAR, 1988):

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$$D = .429C_U + .666C_{Th} + .042C_K \tag{3}$$

where  $C_{Ra}$ ,  $C_{Th}$  and  $C_K$  have the same meaning as in Eq. (1).

The obtained values are shown in Table 3 and range from 3.99 nGy  $h^{-1}$  to 34.79 nGy  $h^{-1}$  with an average of 19.27 nGy  $h^{-1}$ . The mean dose rates are less than the dose rate in air outdoors from terrestrial gamma rays in normal circumstances, 57 nGy  $h^{-1}$  (UNSCEAR, 2000) and less than the world average value of 44 nGy  $h^{-1}$  (UNSCEAR, 1982,1988).

To estimate the annual effective dose in units of  $\mu$ Sv, the conversion coefficient from dose rate to effective dose, 0.7 Sv Gy<sup>-1</sup> and outdoor occupancy factor of 0.2 (UNSCAR, 2000) are used. The obtained values of the annual effective dose rates due to NORM are given in Table 3. The levels range from 4.89  $\mu$ Sv y<sup>-1</sup> to 42.7  $\mu$ Sv and the average is 23.78  $\mu$ Sv and the minimum and the maximum are recorded due to soil samples from L3 and L1, respectively. While the worldwide average annual effective dose is approximately 500  $\mu$ Sv, the results for individual countries are generally within the 300-600  $\mu$ Sv range.

# 4.1.4. The dose rates from <sup>137</sup>Cs

The absorbed dose rate in air at 1 m from the ground surface due to  $^{137}$ Cs levels in soil was calculated using the absorbed dose rate conversion factor 0.1125 nGy h<sup>-1</sup> per Bq kg<sup>-1</sup>. This factor was found on the basis of dose-rate conversion factor  $4.5 \times 10^{-13}$  Gy h<sup>-1</sup> per Bq m<sup>-2</sup> (Leung et al., 1990), and considering a rough soil surface and density of 1.29 g cm<sup>-3</sup> in the upper 20 cm of the soil surface (Arapis et al., 1999) and assuming that  $^{137}$ Cs specific activity of 1 Bq kg<sup>-1</sup> in the first 15 cm of soil surface is equivalent to  $^{137}$ Cs contamination of 250 Bq m<sup>-2</sup> (Barisic et al., 1994). The annual effective dose from  $^{137}$ Cs was deduced using the conversion coefficient from dose rate to effective dose, 0.7 Sv Gy<sup>-1</sup> and outdoor occupancy factor of 0.2. The absorbed dose rates from  $^{137}$ Cs levels in parallel with its annual effective dose are presented in Table 3. The annual effective dose ranges from 0.02 to 8.48  $\mu$ Sv. The minimum and maximum are due to the soil samples taken from L7 and L14, respectively, while the average annual effective dose is 1.53  $\mu$ Sv.

## 4.1.5. The total annual effective dose

The annual effective dose from NORM and  $^{137}$ Cs was estimated for each location (Table 3). The obtained average of the total annual effective dose is 25.4  $\mu$ Sv. The contributions of measured radionuclides in the total annual effective dose are 30% from  $^{238}$ U, 32% from  $^{232}$ Th and 32% from  $^{40}$ K while the contribution of  $^{137}$ Cs is 6%.

# 5. The depleted uranium (DU)

The DU was assessed from the percentage of  $^{235}$ U in the total uranium in the 16 tested samples that showed high levels of  $^{238}$ U. The obtained results are given in Table 5 The range of estimated isotopic abundance of  $^{235}$ U in tested samples is 0.61%0.78% and the mean is 0.66%. By considering the standard deviation which is 0.07% the obtained range of  $^{235}$ U% is very close to its abundance in natural uranium (0.72%). On the other hand, the isotopic composition of DU used by the US Department of Defense as quoted in CHPPM (2000) is

 $^{235}$ U = 0.2% and  $^{238}$ U = 99.8%; therefore, the levels of uranium that were detected in the tested soil could be attributed to the natural type.

Location	U ( $\mu g \ kg^{-1}$ )	$^{235}$ U $\mu g kg^{-1}$ )	$(^{235}U/U)100$
L1	2106.45	15.81	0.75
L4	1645.16	11	0.67
L8	1475.81	9.59	0.65
L9	1564.52	9.78	0.63
L11	1403.23	8.9	0.63
L12	1775	13	0.73
L14	1750	10.8	0.62
L15	1354.84	9	0.66
L18	1403.23	8.5	0.61
L22	1306.46	8.1	0.62
L23	1475.81	10	0.86
L25	1717.74	10.8	0.63
L27	1725.81	11	0.64
L31	1540.321	12	0.78
L32	1491.94	10	0.67
L33	1516.13	9.7	0.64
Ave.	1578.28	10.37	0.66
Max.	2106.45	15.81	0.78
Min.	1306.45	8	0.61
S.D.	231.1	2.3	0.07

Table 5. The total uranium and <sup>235</sup> U concentrations in selected samples from the Musandam Peninsula.

#### 6. Conclusion

The results showed that the levels of  $^{238}$  U,  $^{232}$  Th and  $^{40}$  K are relatively low and uniformly distributed in soil. From a radiological hazard point of view, the Ra<sub>eq</sub> levels indicated that the soil in this region is safe and can be used as construction materials without posing any significant radiological threat to the population. In addition, the radiation hazards index indicates levels less than unity; therefore, the radiation exposure is not significant.

The radiological information gained from this study can be used to help in land utilization for different purposes and to characterize the building materials resource sites

 $^{137}$ Cs levels showed wide variability (0.11-61.40 Bq kg<sup>-1</sup>) in the studied locations and this reveals a great diversity in the properties and textures of tested soil.

The total annual external effective radiation dose from the measured radionuclides is 25.4  $\mu$ Sv. The contributions of radionuclides in the total annual effective dose are 30% from <sup>238</sup>U, 32% from <sup>232</sup>Th and 32% from <sup>40</sup>K while the contribution of <sup>137</sup>Cs is 6%.

The obtained results revealed that the mean of isotopic abundance of  $^{235}$ U is 0.66%; therefore, the uranium detected in the investigated soil is almost of the natural type.

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#### References

Al-Jundia, J., Al-Bataina, B.A., Abu-Rukah, Y. and Shehadeh, H.M., "Natural Radioactivity Concentrations in Soil Samples along the Amman Aqaba Highway, Jordan", Radiation Measurements, 36, 555-560, 2003.

Al-Masri, M.S., "Vertical Distribution and Inventories of <sup>137</sup>Cs in the Syrian Soils of the Eastern Mediterranean Region", Journal of Environmental Radioactivity, 86, 187-198, 2006.

Arapis, G., Chesnokov, A., Ivanova, T., Potapov, V. and Sokolik, G., "Evaluation of Dose Equivalent Rate Reduction as a Function of Vertical Migration of <sup>137</sup>Cs in Contaminated Soils", Journal of Environmental Radioactivity, 46, 251-263, 1999.

Arapis, G.D. and Karandinos, M.G., "Migration of <sup>137</sup>Cs in the Soil of Sloping Semi-Natural Ecosystems in Northern Greece", Journal of Environmental Radioactivity, 77, 133-142, 2004.

Barisic, D., Prohic, E. and Lulic, S., "Some Problems of the Radiometric Mapping of Carbonate Terrain. An Example from Istria (Croatia)", Geology Croatia, 47, 3-82, 1994.

Beretka, J. and Mathew, P.J., "Natural Radioactivity of Australian Building Materials, Industrial Wastes and By-Products", Health Physics, 48, 87-95, 1985.

Bleise, A., Danesi P.R. and Burkart, W., "Properties, Use and Health Effects of Depleted Uranium (DU): a General Overview", Journal of Environmental Radioactivity, 64, 93-112, 2003.

Chikasawa, K., Ishii T. and Ugiyama, H., "Terrestrial Gamma Radiation in Kochi Prefecture, Japan". Journal of Health Sciences, 47, 361-372, 2001.

CHPPM., Health Risk Assessment Consultation No. 26-MF-7555-00D, Depleted Uranium-Human Exposure Assessment and Health Risk Characterization in Support of the Environmental Exposure Report 'Depleted Uranium in the Gulf' of the Office of the Special Assistant to the Secretary of Defense for Gulf War Illnesses, Medical Readiness, and Military Deployments (OSAGWI), OSAGWI Levels I, II, and III Scenarios, 15 September, 2000.

DoAF (Department of the Air Force), Memorandum from Headquarters, Ogden Air Logistics Center, Subject: Gulf War Depleted Uranium Monitions Expenditure, April 30, 1997.

Ebert, D., Hoerner, J., Kang, C., White, B., Biegalski, S. and Massari, J., "Background Spectrometric Measurements in the Vicinity of the Calvert Cliffs Nuclear Power Plant," Transactions of American Nuclear Society Summer Meeting, San Diego, CA, June, 1990.

El-Reefy, H.I., Sharshar, T., Zaghloul, R. and Badran, H.M., "Distribution of Gamma-ray Emitting Radionuclide in the Environment of Burullus Lake: I. Soils and Vegetations". Journal of Environmental Radioactivity, 87, 148-169, 2006.

Goddard, C.C., "Measurement of Outdoor Terrestrial Gamma Radiation in the Sultanate of Oman" Health Physics, 82, 869-874, 2002.

Hamilton, E.I., "Depleted Uranium (DU): a Holistic Consideration of DU and Related Matters", The Science of the Total Environment 281, 5-21, 2000.

Harley, N.H., Foulkes, E.C., Hilborne, L.H., Hudson, A. and Anthony, C.R., "A Review of the Scientific Literature as it Pertains to Gulf War Illnesses", Vol. 7, Depleted Uranium. RAND, Corporation National Defense Research Institute, Washington, USA, 1999.

Hayambu, P., Zaman, M.B., Lubaba, N.C.H, Munsanje, S.S. and Muleya, D., "Natural Radioactivity in Zambian Building Materials Collected from Lusaka", Journal of Radioanalytical Nuclear Chemistry, 199, 229-238, 1995.

Holgye, Z. and Maly, M., "Vertical Distribution and Migration Rates of <sup>239+240</sup> Pu, <sup>238</sup> Pu, and <sup>137</sup> Cs in the Grassland Soil in Three Locations of Central Bohemia", Journal of Environmental Radioactivity, 47, 135-147, 2000.

Leung, K.G., Lau, S.Y. and Poon C.B., "Gamma Radiation Dose from Radionuclide in Hong Kong Soil", Journal of Environmental Radioactivity, 11, 179-290, 1990.

Nada, R.M., Mirjana, M.M., Dargana, J.T., Mirjana, R.C., Dusan D.G., Milam, P.O., Dragan S.V. and Rade, N.B., "Uranium Content in the Soil of the Federal Republic of Yugoslavia after NATO Intervention", Archive of Oncology, 9, 245-249, 2001.

Obed, R.I., Farai, I.P. and Jibiri, N.N., "Population Dose Distribution Due to Soil Radioactivity Concentration Levels in 18 Cities across Nigeria", Journal of Radiological Protection, 25, 305-312, 2005.

OECD, Organization for Economic Cooperation and Development, "Exposure to Radiation from the Natural Radioactivity in Building Materials", Report by a Group of Experts of the OECD Nuclear Energy Agency, OECD, Paris, France, 1979.

Orabi, H., Al-Shareaif, A. and El Galefi, M., "Gamma-ray Measurements of Naturally Occurring Radioactive Sample from Alkharje City", Journal of Radioanalytical Nuclear Chemistry, 269, 99-102, 2006.

Papachristodoulou, C.A., Assimakopoulos, P.A., Patronis, N.E. and Ioannides, K.G., "Use of HPGe  $\gamma$ -ray Spectrometry to Assess the Isotopic Composition of Uranium in Soils", Journal of Environmental Radioactivity, 64, 195-203, 2003.

Price, K.R., "The Depth Distribution of <sup>90</sup>Sr, <sup>137</sup>Cs, <sup>239+240</sup>Pu in Soil Profiles Samples", Radiochimica Acta, 54, 145-147, 1991.

Rani, A. and Singh, S., "Natural Radioactivity Levels in Soil Samples from Some Areas of Himachal Pradesh, India Using  $\gamma$ - ray Spectrometry", Atmospheric Environment, 39, 6306-6314, 2005.

Ritchie, J.C. and McHenry, J.R., "Application of Radioactive Fallout Cesium-137 for Measuring Soil Erosion and Sediment Accumulation Rates and Patterns: a Review", Journal of Environmental Quality, 19, 215-233, 1990.

Rodrigue, J.P., "Straits, Passages and Chokepoints. A Maritime Geostrategy of Petroleum Distribution", Les Cahiers de Geographie du Quebec 48 (135), 357-374, 2004.

Sigurgeirsson, M.A., Arnalds, O., Palsson, S.E., Howard, B.J. and Gudnason, K., "Radiocaesium Fallout Behaviour in Volcanic Soils in Iceland", Journal of Environmental Radioactivity, 79, 39-53, 2005.

Staunton, S., Dumat, C. and Zsolnay, A., "Possible Role of Organic Matter in Radiocaesium Desorption in Soils", Journal of Environmental Radioactivity, 58, 163-173, 2002.

Tso, M.Y. and Leung, J.K., "Population Dose Due to Natural Radiations in Hong Kong", Health Physics 8, 555-78, 2000.

U.S. E.I.A., (United States Energy Information Administration Independent Statistical and Analysis). World Oil transit Checkpoints. Country Analysis Briefs, 2008.

UNSCEAR, (United Nations Scientific Committee on the Effects of Atomic Radiation), Report to the General Assembly United Nations, New York, 1982.

UNSCEAR, (United Nations Scientific Committee on the Effects of Atomic Radiation), Report to the General Assembly United Nations, New York, 1988.

UNSCEAR, (United Nations Scientific Committee on the Effects of Atomic Radiation), Report to the General Assembly, United Nations, New York, 1993.

UNSCEAR, (United Nations Scientific Committee on the Effects of Atomic Radiation), Report to the General Assembly United Nations, New York, 2000.

UNSCEAR, (United Nations Scientific Committee on the Effects of Atomic Radiation), Report to the General Assembly United Nations, New York, 2008.

Yu, K.N., Guan, Z.J., Stoks, M.J. and Young, E.C., "The Assessment of Natural Radiation Doses Committed to the Hong Kong People", Journal of Environmental Radioactivity, 17, 31-48, 1992.