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Radiological Assessment and Mechanical Separation of NORM Contaminated Soil from Iraqi Oil Fields

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Abstract

Naturally occurring radioactive materials (NORM) contaminated sites at Al-Rumaila Iraqi oil fields have been characterized as a part of soil remediation project. Activity of radium isotopes in contaminated soil have been determined using gamma spectrometer High Purity Germanium detector (HPGe) and found to be very high for Al-Markezia, Al-Qurainat degassing stations and storage area at Khadhir Almay region. The activity concentration of samples ranges from 6474.11±563.8 Bq/kg to 1232.5±60.9 Bq/kg with mean value of 3853.3 Bq/kg for ^{226}Ra, 843.59±8.39 Bq/kg to 302.2±9.2 Bq/kg with mean value of 572.9 Bq/kg for ^{232}Th and 294.31±18.56 Bq/kg to 156.64±18.1 Bq/kg with mean value of 225.5 for ^{40}K. Six hazard indexs radium equivalent, representative level index, adsorbed dose rate in air, annual effective dose equivalent, external hazard index, and internal hazard indexes were calculated to estimate the potential radiological health risk in soil and dose rate associated with it and found to be high. Screening of contaminated soil was performed to evaluate the feasibility of particle size separation. The fractions obtained varied between 75 µm (200 mesh) to 300µm (48 mesh).The results show that the largest weight percent in fine particle size cut (-75, -125+75, -250+125) µm is 73.9% and all radium isotopes are concentrated in 37.5µm particle size while small fluctuations are observed in the other particle size cuts.

Key Words: NORM, Radium isotopes, radiological assessment, mechanical separation

Introduction

Radium is the heaviest alkaline earth metal belonging to Group IIA of the periodical table. It has 25 isotopes with mass numbers between 206 and 230, all of them are radioactive. The most abundant among the naturally occurring isotopes are ^{226}Ra with a half-life of 1620 years from the uranium series (^{238}U), and ^{228}Ra with a half-life of 5.8 years from thorium series (^{232}Th). These two isotopes of radium are also the most radiotoxic and very significant from a radiological protection viewpoint due to their relatively presence in nature,

long half-lives, and high dose conversion factors. In the 1950s radium was identified as a pollutant of the environment, caused by uranium mining and milling in the USA. Other, non-nuclear branches of industry have also been identified as significant sources of environmental pollution due to radium release as– phosphate fertilizer production, oil and gas exploitation, underground mining of different raw materials (heavy sands, coal, gold etc.).[1]

Naturally occurring radioactive materials (NORM) containing ^{232}Th ^{238}U- series nuclides can be accumulated and concentrated in surface equipment and tubing in the form of sludge and scale as a consequence of chemical and physical processes associated with the oil and gas industry [2]. In addition, produced water (brine water) associated with oil is typically separated from oil and disposed of by one of these methods, like down an injection well or discharged into the environment for evaporation [3].

In some cases, produced water amount is greater than the amount of oil produced. Therefore, it may be considered the largest volume of radioactive waste produced by the oil and gas industry [2]. Most Middle East operating companies dispose their wastes water into unlined pits and lagoons. Subsequently, the projection water is drained to underground leaving radioactive precipitate within the soil that finally required remedial or treatment action in accordance with radiation protection principles. Therefore, the remediation projects to take away and treat contaminated soil have been started in order to reduce the hazard to workers and public. [2,4]. In Iraqi oil field (Rumaila), produced water is discharged into the environment for evaporation. Uncontrolled disposal of this type of waste could lead to pollute the environmental and, therefore, finally lead to radiation exposure of workers in this field and members of the public.

The goal of the present study is the assessment of the radiation exposure of the existing contamination of land areas from oil and gas industry in Iraqi oil fields. Radiological assessment for Al-Markezia and Al-Qurainat degassing stations and, mechanical separation are made to reduce the cost of disposal by reducing the volume of NORM contaminated soil.

Experimental Work

1- Mechanical Separation

Three different soil samples from Al- Markezia and Al-Qurainat Degassing stations and Khadhir almay region were identified for radiological assessment. These soil samples are mixed and separated for different particle size ((+300), (-300+250), (-250+125) , (-125+75), (-75)) using the test sieve shaker (Impact, SV003). The material held on each of the sieves or the fraction of each particle size was separated, collected, weighed, and percentage of each weight fraction was calculated. The analysis was also carried out regarding average size, mass fractions, and cumulative mass fraction. This step was made in laboratories of Radiological and Nuclear Safety Directorate (RNSD) / Al-Tuwaitha site / Ministry of Science and Technology.

2- Sample Preparation

The sample preparation depends on type and quantity of samples under investigation. The soil samples were dried and then moved to spatial container. 100 or 200 ml of each soil sample was placed in standard plastic container then was sealed and stored for 3-4 weeks prior to measurement to permit the decay daughter ^{214}Bi, ^{214}Pb,

^{212}Bi, ^{212}Pb, ^{228}Ac to establish an equilibrium with ^{226}Ra, ^{228}Ra and ^{224}Ra and counting by Gamma spectrometer[5]. The analytical measurements were made in the laboratories of the Environmental Radiation Control Department at the Lebanese Atomic Energy Commission (LAEC) in Lebanon.

3- Measurement of Sample Particle Size

Radionuclide activity concentration was analyzed using Gamma spectrometer from Canberra equipped with extended range low-level coaxial High Purity Germanium (HPGe) detector with high resolution (2.1 keV at 1332 keV) and 50% relative efficiency was utilized in this study. In order to reduce the background radiation, the detector was surrounded with a 10-cm-thick lead shield and by a 0.5 cm copper layer to attenuate the X-rays emitted by the lead shield. The detector was linked to standard integrated data processor DSA 1000 desktop inspector electronics from Canberra and the spectra were accumulated in 8K MCA.

A standard multigamma radioactive source from Isotope Products Laboratories (ISO) used to the energy calibration. This was done by preparation standard sample in the same geometry as the samples to be analyzed, this step occurred once in week or when needed, the efficiency calibration is estimated. Moreover, efficiency curves were corrected for attenuation and absorption. The background spectra were measured regularly under the same conditions used for the sample and applied to correct the calculated sample activities. The linearity and the resolution of the detector were checked using a standard ^{152}Eu point source. The time counted for each sample ranged between 3 to 48 hr and spectra were analyzed off-line using Genie 2000 software from Canberra Version V3.1.a, including peak search, nuclide identification, activity and uncertainty calculation, and MDA calculation modules.[5]

The method allowed to determine NORM nuclides as summarized in table (1) ^{226}Ra activities was calculated at 186.2 keV after correction for ^{235}U [7]. If interference of the 186.2 keV photon from ^{235}U cannot be excluded, ^{226}Ra activities in the samples were then measured by determining its gamma emitted daughters (^{214}Pb or ^{214}Bi) after equilibrium[6]. ^{228}Ra was determined from the gamma line of its daughter ^{228}Ac at (911.2 keV), as well as ^{224}Ra was determined from gamma line of its daughter ^{212}Pb, and ^{212}Bi at (238.63, and 727.33) respectively because the emanation rate of the ^{226}Ra progeny ^{222}Rn from scales and sludges is typically very low, ^{226}Ra may be measured directly by its γ energy 186.2 keV in all samples with low concentrations of uranium [7].

Table 1, Summary of Gamma spectrometry on NORM counting solids [2]

isotope	Isotopes heuristics	Energy (keV)	Gamma Emissions probability (%)
^{226}Ra	^{226}Ra	186.2	3.56
	^{214}Pb	351.93	35.6
	^{214}Bi	609.31	45.49
^{228}Ra	^{228}Ac	911.2	26.2
^{224}Ra	^{212}Pb	238.63	43.6
	^{212}Bi	727.33	6.65

The two radium isotopes identified to be of primary concern are ^{226}Ra and ^{228}Ra because of potential carcinogenic impact.

Results and Discussion

1- Hazard Assessment for Soil Sample

It is reasonable to develop as many as possible identified radiation health hazard indices analysis to obtain effective conclusion on the human health and environment. Six values have been calculated to assess the radiation health hazards associated with the soil samples as described below [8&9].

Calculation of Radium Equivalent Activity

The distributions of ^{226}Ra, 232Th, and ^{40}K in samples are not uniform therefore, to represent their specific activities by a single quantity to take into calculation the radiation hazards related with them. To define Ra_{eq} activity, it can be assumed that 1 Bq/kg of ^{226}Ra, 0.7 Bq/kg of ^{232}Th or 13 Bq/kg of ^{40}K give the same dose of gamma ray. Radium equivalent has been used as radiological index (Ra_{eq}) in Bq/kg which is calculated using Eq. (1) [10]

$$Ra_{eq} = C_{Ra} + 1.43C_{Th} + 0.077C_K \leq 3 \quad \ldots (1)$$

Where

C_{Ra}: The activity concentrations of 226Ra in Bq/kg

C_{Th}: The activity concentrations of 232Th in Bq/kg

C_K: The activity concentrations of 40K in Bq/kg

Calculation of Representative level index (I_γ)

The representative level index (I_γ) is the second hazard index used in this study for the calculation of gamma radiation related with the natural radioactive materials in the soil. It is calculated using Eq. (2). The safety value for this index is ≤ 1 [11]

$$I_\gamma = \frac{C_{Ra}}{150} + \frac{C_{Th}}{100} + \frac{C_K}{1500} \leq 1 \quad \ldots (2)$$

Calculation of air absorbed radiation dose rate

Gamma radiation effects are usually expressed in terms of adsorbed dose rate in air. At a height of about 1 meter above the ground surface the external terrestrial absorbed dose rate of γ-radiation in air was calculated for ^{226}Ra, ^{232}Th and ^{40}K radionuclides, using Eq. (3). The conversion factor of 0.462 nGy h^{-1}/Bq kg^{-1} for ^{226}Ra, 0.621 nGy h^{-1}/Bq kg^{-1} for ^{232}Th, and 0.0417 nGy h^{-1}/Bq kg^{-1} for ^{40}K, equilibrium is assumed between ^{226}Ra and ^{232}Th series with all their daughter and the effect of ^{90}Sr, ^{137}Cs, and ^{235}U decay series can be neglected because of their small contribution to the whole dose from background. [11]

$$D(\text{nGy h}^{-1}) = 0.462C_{Ra} + 0.621C_{Th} + 0.0417C_K \quad \ldots (3)$$

Calculation of Annual Effective Dose

The annual effective dose equivalent outdoor predictable to be absorbed by the human due to the radioactivity in soil was calculated using Eq. (4). To convert absorbed rate in air to effective dose using a conversion factor of 0.7 Sv Gy^{-1}, with an outdoor occupancy factor of 20 and 80% for indoor [11].

$$Outdoor(nSv/y) = (Absorbed\ dose)nGyh^{-1} \times 8760 \times 0.2 \times 0.7\ SvGy^{-1} \quad \ldots (4)$$

Calculation of External and Internal Hazard Index [10]

The external hazard index (H_{ex}) is widely used to reflect to external exposure and can be calculated by Eq. (5):

$$H_{ex} = \frac{C_{Ra}}{370} + \frac{C_{Th}}{259} + \frac{C_K}{4810} < 1 \quad \ldots (5)$$

And the internal hazard indexes (H_{in}) used to reflect to internal exposure to radon and its daughter

$$H_{in} = \frac{C_{Ra}}{185} + \frac{C_{Th}}{259} + \frac{C_K}{4810} < 1 \quad \ldots (6)$$

In order to keep the radiation hazard to be insignificant the value of external and internal radiation hazard index must be less than unity.

The data in Table (2) are summarized of measurements of natural radionuclide (^{226}Ra, ^{232}Th, and ^{40}K) concentration in the collected soil samples from Markezia (M1and M2) and Qurainat (Q1) degassing station, and Khidhr-almay (KH1) where mean value of samples (S7 to S11) were taken. The world average concentrations for ^{226}Ra, ^{232}Th, and ^{40}K in soil sample are 35, 30, 400 Bq/kg respectively. Table (2) shows that, The activity concentration of ^{226}Ra and ^{232}Th in selected soil of degassing station are higher than the world value reported in [10] and these concentration are more than 150 and 44 times for ^{226}Ra and ^{232}Th respectively. The concentration for ^{40}K is lower as compared with the world figures.

The value of radium equivalent activity, representative level index, absorbed gamma radiation dose, annual effective dose equivalent outdoor, internal and external hazard index are shown in Table (3). Radium equivalent is calculated from Eq. (1) values of contaminated samples range from 7593.83Bq/kg (Markezia) to 1679.93Bq/kg (Qurainat) with mean value 4636.88 Bq/kg which is higher than the safe limit (370 Bq/kg) recommended by Organization for Economic Cooperation and Development (OECD)[8]. The recommended value of annual effective dose equivalent is 1 mSv/year for the personal of the public and 20 mSv/year for the workers in the radiation field. This is fixed by the International Commission on Radiological protection (ICRP). From table (3) it is clear that the absorbed dose rate calculated by Eq. (3) ranges from 3478.59 (Markezia) to 765.36 (Qurainat) nGyhr^{-1} with an average value of 2121.98 nGyhr^{-1}. The world wide average annual effective dose is approximately 0.5 mSv. The annual effective dose of these samples is higher than the acceptable value except Q1. The values of internal and external hazard index are higher than unity therefore, according to the report of radiation protection 112; the soil isn't safe and can't be used as a building material without any significant radiological hazard to population [9].

2- Particle Size Distribution

Particle size measurement was made using sieve analysis.After sieving, each particle size cut was collected, weighted and measured.

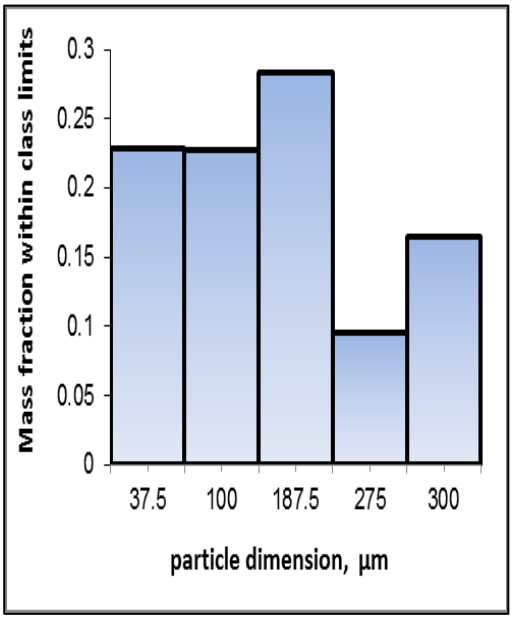

Fig. 1, Histogram presentation of screen analysis

The results are shown in Table (4) and Figure (1 and 2), plotted as a histogram, and cumulative distribution.

The fractional or acumulative distribution curves are made by assuming the material between two screens to have a particle diameter that is the arithmetic average of the two screen openings.

Table 2, the activity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K in Bq/kg measured in contaminated soil

Sample No.	A.Conc. of ^{226}Ra Bq/kg	A.Conc. of ^{232}Th Bq/kg	A.Conc. of ^{40}K Bq/kg
M1	6474.11±563.8	774.59±9.19	156.64±18.1
M2	4073.91±141.12	843.59±8.39	170.05±13.01
Q1	1232.5±60.9	302.2±9.2	198.5±14.6
KH1	3619.4±12.9	843.3±9.9	294.31±18.56
S7-S11	5508.3±217.6	1363±69.2	315.5±24.5

Table 3, calculated values of hazard assessment

Sample No.	Radium equ. activity (Bq/kg) Ra_{eq}	Representative level index (Iγ)	Absorbed dose rate D (nGy hr^{-1})	Annual effective dose rate mSv AEDE	External hazard index H_{ex}	Internal hazard index H_{in}
M1	7593.83	51.01	3478.59	4.27	20.52	38.02
M2	5293.34	35.71	2413.11	2.96	14.30	25.31
Q1	1679.93	11.37	765.36	0.94	4.54	7.87
KH1	4847.98	32.76	2208.12	2.71	13.10	22.88
S7-S11	7481.95	50.56	3404.52	4.18	20.22	35.10

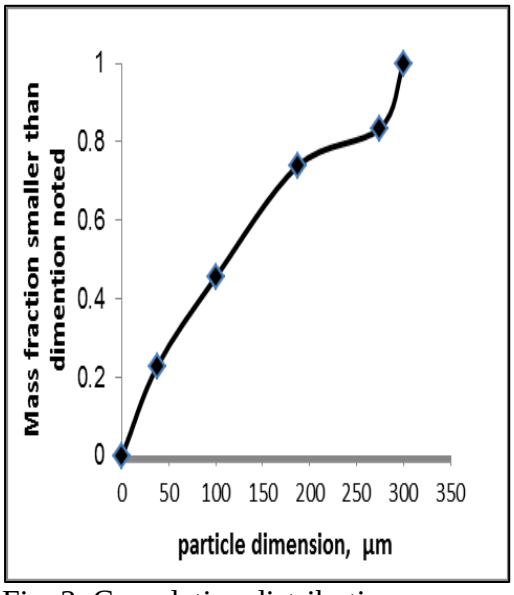

Fig. 2, Cumulative distribution curve

The results were obtained for five fractions with different mesh size and their weight percent in the samples. The fractions obtained varied between 75 µm (200 mesh) to 300µm (48 mesh). Figures above show that the largest weight percent in fine particle size (-75, -125+75, -250+125) µm is 73.9% . The average diameter d_{50} was determined from Fig(2) and was equal to112 µm. The surface mean diameter of soil samples was calculated from Eq. (7) and found to be equal to 48 µm. This value ensures the high contribution of the fines in the soil sample, considering the surface mean in the calculations.

$$d_{50} = \sqrt{\frac{\sum_{i=1}^{k} \frac{x_i}{D_p}}{\sum_{i=1}^{k} \frac{x_i}{(D_p)^3}}} \quad \ldots (7)$$

Radioactivity Distribution with Soil Particle Size

Each particle size soil sample was analyzed for the exposure dose rates by portable instrument to select the dealing method. The results are presented in table (5).

It can be seen that high values of exposure rate for all samples are observed, and greater than three times of the background (BG) value, therefore, personal protection equipment was used. Furthermore the work area was carefully prepared to reduce the contamination of the area and equipments

Table 4, Results of typical screen analysis

Sample No.	Mesh No.	particle size µm	Mean particle size D_p (µm)	Weight of sample (g)	Weight %	Cumulative
S1	-200	-75	37.5	544.8	22.8	0.228266
S2	-115 + 200	-125 +75	100	543	22.8	0.455778
S3	- 60 + 115	-250 +125	187.5	676.6	28.3	0.739267
S4	- 48 + 60	-300 +250	275	228	9.6	0.834796
S5	48	300	300	394.29	16.5	1

Table 5, Exposure rate measurement for soil samples

Sample No.	particle size µm	Mean particle size µm	Exposure rate nSv/hr
S1	-75	37.5	677
S2	-125 +75	100	397
S3	-250 +125	187.5	313
S4	-300 +250	275	225
S5	300	300	393
BG			60

Table 6, Activity concentrations for radium isotopes and potassium of contaminated soil for Particle size distribution

Sample No.	A.Conc. of ^{226}Ra Bq/kg			A.Conc. of ^{228}Ra Bq/kg	A.Conc. of ^{224}Ra Bq/kg		A.Conc. of ^{40}K Bq/kg
	186.2 keV	351.93 keV	609.31 keV	911.2 kev	238.63 keV	727.33 keV	1460 keV
S1	14520±279.2	13290±222.8	12850±211	3367±116.5	3935±81.7	4545±75.4	245.5±9
S2	5759±119.2	5398±91.4	5202±86.5	1549±55	1751±36.9	1984±43.6	339.8±20.7
S3	5160±208.9	4694±188.1	4330±174.2	1234±62.2	1503±62.6	1623±66.3	332.8±17.7
S4	7038±289.9	6267±251.5	5714±230.4	1672±85.5	2072±86.6	2336±101	240.9±39
S5	7669±313	6903±276.8	6344±255.5	2007±101.7	2407±100.4	2596±108.7	223±43

After that all these samples were measured by gamma spectrometer techniqe and the results are shown in table (6). ^{226}Ra, ^{228}Ra, and ^{224}Ra activities were determined in different particle size soil samples from the contaminated area in al-Rumaila oil field.

The results of samples are presented in table (6) and Fig. (3, 4, and 5). It was observed that all radium isotopes are concentrated in 37.5µm particle size and small fluctuations in other particle size. Furthermore, The activity concentrations of radium isotopes (^{226}Ra, ^{228}Ra, and ^{224}Ra) with small particle size is higher than the activity concentration with large particles. For that, determination of radium isotopes distribution with particle size is more important for volume reduction of radioactive waste [12].

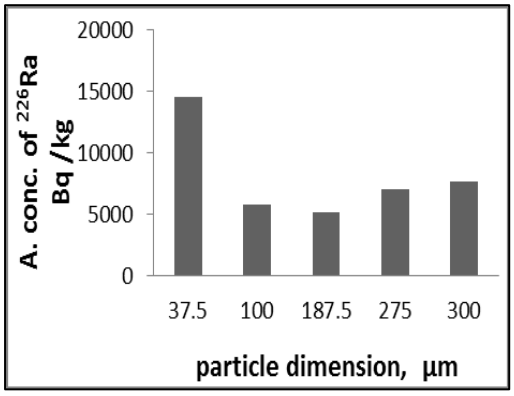

Fig. 3, Particle Size distribution with Activity Concentrations for ^{226}Ra

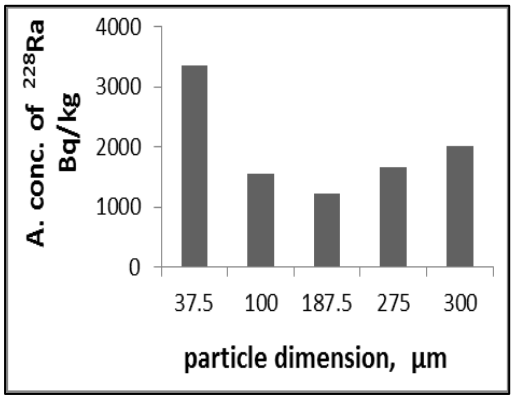

Fig. 4), particle size distribution with activity concentrations for ^{228}Ra

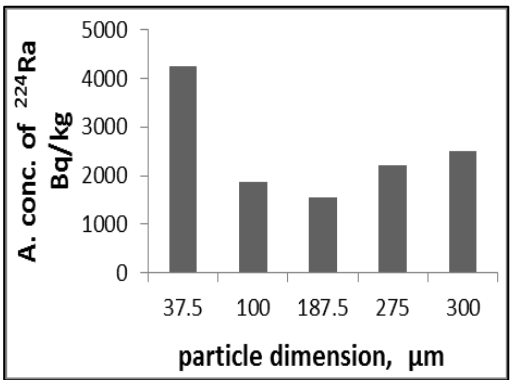

Fig. 5, particle size distribution with activity concentrations for ^{224}Ra

3- Evaluation of the Homogeneity of the Contaminated Soil

To insure that all contaminated soil was completely homogenized the five random samples were taken, as shown in Fig. (6).

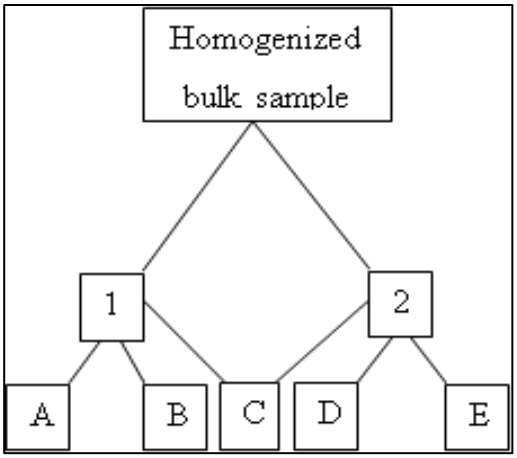

Fig. 6, Scheme for the Preparation and Homogeneity Test

The analytical results of the homogenized contaminated soil sample of ^{226}Ra, ^{228}Ra, ^{224}Ra, and ^{40}K are shown in table (7) & Figure (7, 8, 9 and 10). The mean value for ^{226}Ra, ^{228}Ra, ^{224}Ra, and ^{40}K are 6088, 1363.4, 1785.8, and 315.46 Bq/kg with standard deviation as 138.51, 18.28, 30.69, and 32.6 respectively. The results show that the higher standard deviation occurs for ^{40}K while the standard deviation for ^{226}Ra, ^{228}Ra, ^{224}Ra is approximately the same for that the aggregate may be regarded as

homogenous because all values were distributed on the mean values.

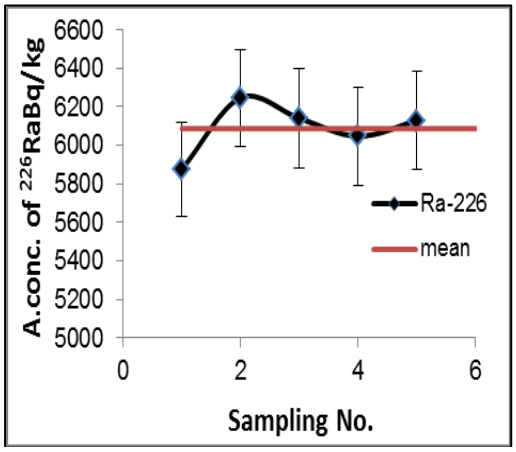

Fig. 7, Representation of homogeneity test results for ^{226}Ra concentration in contaminated soil

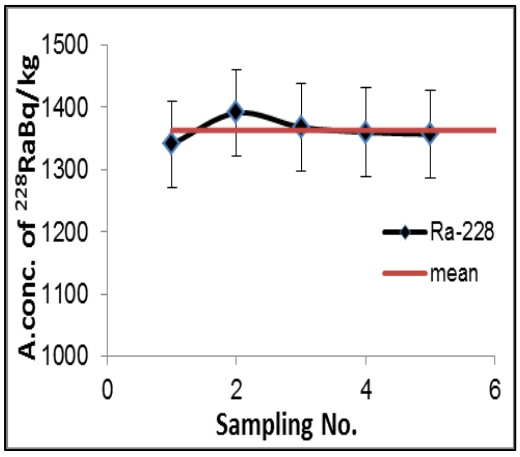

Fig. 8, Representation of homogeneity test results for ^{228}Ra concentration in contaminated soil

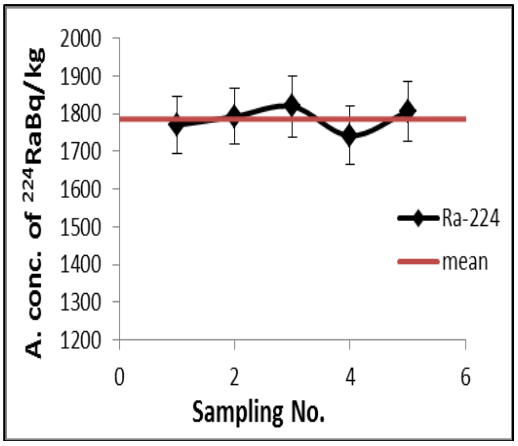

Fig. 9, Representation of homogeneity test results for ^{224}Ra concentration in contaminated soil

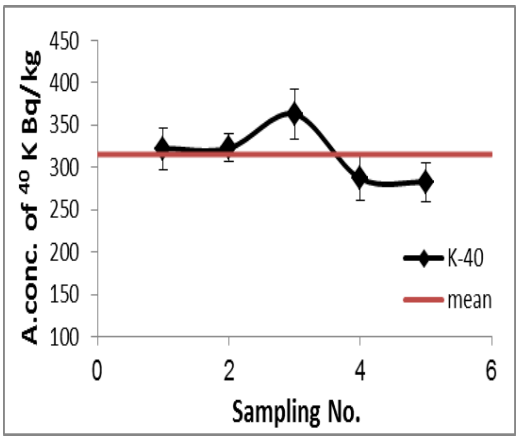

Fig. 10, Representation of homogeneity test results for ^{40}K concentration in contaminated soil

Table 7, Activity concentration for radium isotopes and potassium in homogenized contaminated soil

Sample No.	A.conc. of ^{226}Ra Bq/kg			conc. of ^{228}Ra Bq/kg	A.conc. of ^{224}Ra Bq/kg		.conc. of ^{40}K Bq/kg
	186.2 keV	351.93 keV	609.31 keV	911.2 keV	238.63 keV	727.33 keV	1460 keV
S7	5875±244.3	5213±209.9	4918±198.6	1341±69.2	1655±69.3	1885±84.5	322.3±24.5
S8	6247±253.1	5562±222.8	5110±205.6	1391±70.2	1721±71.7	1864±76.5	322.7±16.5
S9	6141±257.6	5493±220.9	5087±205.7	1368±71.2	1722±72.3	1916±88.7	362.9±29
S10	6048±253.7	5362±215.7	4926±199.2	1360±70.7	1668±70.1	1815±84.6	286.9±25.9
S11	6129±253.8	5488±220.4	5021±202.6	1357±69.8	1713±71.7	1899±84.2	282.5±22.6

Conclusions

1- The activity concentrations of radium isotopes (^{226}Ra, ^{228}Ra, and ^{224}Ra) with small particle size are higher than the activity concentration with large particles.
2- The activity concentration of ^{226}Ra and ^{232}Th in selected soil of degassing station are higher than the world value reported in [10] and these concentrations are more than 150 and 44 times for ^{226}Ra and ^{232}Th respectively. While the concentration for ^{40}K is lower as compared with the world figures.
3- The largest weight percent in fine particle size (-75, -125+75, -250+125) µm is 73.9% .

Nomenclature

AEDE	Annual effective dose rate
C_{Ra}	The activities concentration of 226Ra
C_{Th}	The activities concentration of 232Th
D	Absorbed dose rate of γ-radiation in air
d50	Mean particle diameter
H_{ex}	The external hazard index
H_{in}	The internal hazard indexes
Iγ	The representative level index
A.conc	Activity concentrations
IAEA	International Atomic Energy Agency
KH1	Soil sample from Khidhr-almay region
M1,M2	Soil samples from Markezia degassing station
MCA	Multi channel analyzer
MOST	Ministry Of Science and Technology
NORM	Naturally occurring radioactive materials
Q1	Soil sample from Qurainat degassing station
S1,S2,,S11	Soil samples

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