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Radiometric properties of virgin and cultivated soil around the Shazand Refinery Complex in Iran

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► ***Original article***

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ABSTRACT

Background: One of the main sources of exposure to radiation is terrestrial radionuclides in the environment. These radioisotopes are present in the Earth's crust and can be increased by human activity such as mining of coal, oil, and minerals. ***Materials and Methods:*** In this study, 39 soil samples including virgin and cultivated were collected from around of the Shazand Refinery Complex(SRC) using a template and experimental method. The gamma spectrometry method was used to measure the specific activity of the ^{226}Ra, ^{232}Th, ^{40}K and ^{137}Cs radionuclides. ***Results:*** The average concentrations of ^{226}Ra, ^{232}Th, ^{40}K and ^{137}Cs in the cultivated (virgin) soil samples were 21.95 ± 0.27 (23.99 ± 0.37), 25.37 ± 0.29 (31.74 ± 0. 38), 416.72 ± 1.88 (461.09 ± 2.68) and 5.13 ± 0.08 (5.51 ± 0.14) in Bq kg^{-1}. Excess lifetime cancer risk (ELCR) was obtained for cultivated (virgin) soil as 0.19 × 10^{-3} (0.22 × 10^{-3}), which is close to the world average (0.29 × 10^{-3}) and lowers than the maximum acceptable value (10^{-3}). ***Conclusion:*** The specific activities of natural radionuclides were in the global range and are lower than the maximum allowable value. The distribution map of ^{226}Ra and ^{137}Cs indicated some part of ^{226}Ra, distributed as fly ash from a chimney, caused by the incineration of refinery waste. The radiological parameters calculated for both types of soil were lower than the maximum admissible values, and therefore there is no radiological hazard for people living in this area.

Keywords: *Dosage, radionuclides, radiation, soil, pollution.*

INTRODUCTION

A high percentage of total environmental irradiation results from natural radiation [1]. In particular, natural radioactive nuclei in soil, water and air are responsible for human exposure to radiation [2]. Natural radioactive nuclei easily move in the human environment and their mobility results from their geochemical properties [3]. Radionuclides (uranium, thorium series and potassium) are found naturally in soil, stones and water. The average amount of uranium and thorium in the earth's crust is estimated to be 2.7 mg kg^{-1} and 9.6 mg kg^{-1}. Potassium is one of the main components of the earth's crust, creating 2.8 percent of the shell composition [4]. Human activities, such as mining (ore crushing), metal extraction processes, nuclear fuel production, and end-of-fuel products, are transmitted to the environment to a significant extent by radioactive nuclei [1]. In recent years, concentrations of natural radionuclide in various petroleum and oils have been measured in many countries [5, 6]. Part of the radionuclides in the oil is burned during the refinery process along with the waste distillation tower and dispersed through the chimneys in the air, and after cooling; the ash contains radioactive nuclei, falls on the surrounding soil and increases the radiation level. It depends primarily on the ambient temperature, humidity of air and wind direction. Man-made radionuclides are produced from nuclear industrial activities, nuclear power plant accidents, or military uses. Radionuclides produced from nuclear activities

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are dispersed in ecosystems of the environment depending on their physicochemical properties and conditions prevailing in the environment [1]. According to the report by United Nations Scientific Committee on the effect of Atomic Radiation, the atmospheric nuclear test of nuclear weapons from 1945 to 1980 and nuclear accidents such as the Chernobyl (1986) nuclear reactor are the main sources of environmental pollution by ^{137}Cs and ^{90}Sr radionuclides. The accident in Chernobyl released 3.8×10^{16} Bq of radionuclide ^{137}Cs in atmosphere [1]. Every nuclear explosion causes the uncontrolled release of a significant amount of radioactive materials into the atmosphere, which are gradually stored all over the world. ^{90}Sr with a half-life of 28.78 years is considered as an important health hazard because it replaces calcium in the bone. Among the class of artificial isotopes released as fission products, ^{137}Cs is the most prominent isotope detected by its gamma radiation on the earth's surface [7]. ^{137}Cs penetrates the soil through water step by step and can enter to groundwater. This cesium remains for long time in the upper layers of the soil and joins the soil particles. The aim of the study was to investigate the impact of radioactive pollutant outlets from refinery chimneys on cultivated and virgin land around these complexes. Also in this work, the concentration of radionuclides in cultivated and virgin soil was measured, as well as to assess radiological parameters such as the absorbed dose in the air, the annual effective dose (internal and external), internal and external hazard indices and the gamma index from the point of impact on public health living in these areas.

MATERIALS AND METHODS

Introducing areas under study

Shazand refining and petrochemical plants are located in the city of Shazand in Markazi province in Iran at kilometer 22 of the Arak-Khoramabad road and have been built on an area of 523 hectares. One of the largest producers of oil components, polymers and chemical products, as well as one of the most important projects in Iran was built in 1992[8]. There are several chimneys that spread fly ash from combustion to the environment.

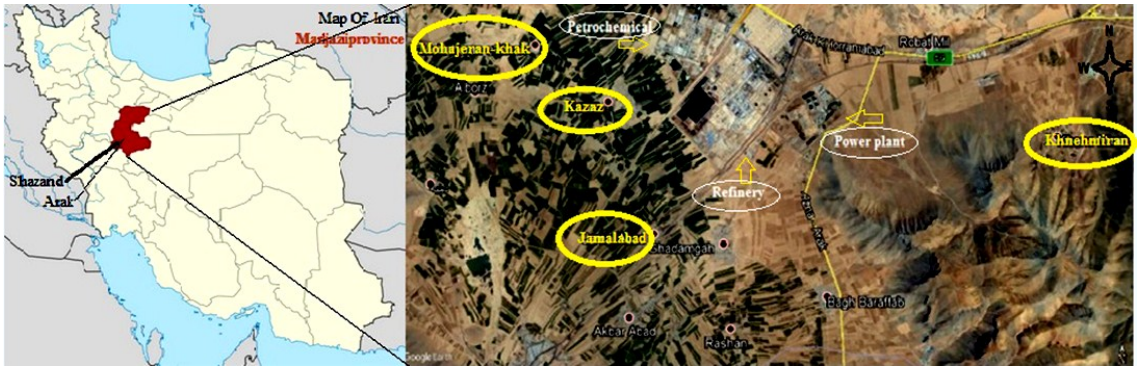


Figure 1. Sampling location map showing study area, Khanemiran, Mahajeran-khak, Kazaz and Jamalabad around of SRC.

Sampling and sample preparation

In this study, 39 soil samples were collected from virgin and cultivated areas in Kazaz, Mohajeran, Jamal Abad and Khane-Miran in fields in the Markazi province of Iran to measure the concentration of radionuclides. 9, 8, 12 and 10 samples were prepared respectively from these areas. Figure 1 shows the location of studied area. A random and experimental sampling method was used to collect samples. Of all samples, stones, roots and waste were removed. After the drying and milling process, the samples were sieved by a 0.508 mm pore size sieve and prepared for each homogeneous sample. Samples packed in cylindrical containers, weighing 300 grams and sealed with

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silicone glue to prevent escape of radon gas (creating an equilibrium between ^{226}Ra and their daughters in the sample [9].

Gamma spectrometry and analysis

A high purity germanium (HPGe) P-type coaxial detector (GCD30195BSI model manufactured by Baltic Scientific Instrument LTD, 005- Latvia) was used to measure gamma spectra of soil samples. The energy resolution for 1332 keV (^{60}Co) and the relative efficiency of detector were 1.95 keV and 30% respectively. Detector and preamplifier were shielded in a chamber of three layers composed of 10 cm thick lead, 1.5 mm thick cadmium and 2 mm thick by copper to reduce background radiation [10]. Multi-Channel Analyzer (8 k MCA) and with other electronic accessories were in the bin set and connected to PC. Initially the detector was calibrated so that the energy of each peak should be in accordance with corresponding channel number. The energy calibration was performed using ^{241}Am, ^{152}Eu and ^{137}Cs sources. To register the spectrum of each sample, the packed soil samples were placed face to face of detector in center of shield chamber individually. The time of spectrometry depends on the efficiency of the detector (vice versa) and it must be long enough for the photo peaks to have a Gaussian shape. The soil samples spectrum was registered for 86400 seconds and recorded by the Lsrmbsi software (Baltic Scientific Instrument LTD, 005-Latvia). Calibration of the system was carried out using a standard soil source, which contained ^{152}Eu, ^{241}Am, ^{133}Ba and ^{137}Cs. Gamma Ray reference material from uranium, thorium and potassium (RGU-1, RGTh-1, RGK-1), which was prepared by the International Atomic Energy Agency (IAEA), was used to control the quality of efficiency calibration in the energy range 58.78 - 2478, 09 keV. The measurement results of reference material are shown in table 1. To evaluate the uncertainty of measurement amount of reference material, calculated Z-score value [11]

The z-score was calculated using equation 1:

$$Z = \frac{e-c}{\sqrt{u_e^2+u_c^2}} \quad (1)$$

Where; e is the experimental result that has been measured, c is the certified value reported by the IAEA, u_e, u_c are the uncertainties of the measured and reported values respectively. The acceptable **Z** value for the measured activity should be | Z | <2. Standard deviations higher or lower than the certified value are determined by z-score [11].

Table 1. Certificated and measured values for reference material IAEA and z-score values.

Reference material	Contains elements	certified value (Bq kg $^{-1}$)	Average activity (Bq kg $^{-1}$)	measured activity (Bq kg $^{-1}$)	Z - score
RGU-1	7.09% U (uranium ore)	4910 - 4970	4940± 30	4979.54± 30.78	0.92
RGTh-1	2.89% Th (thorium ore)	3160- 3340	3250± 90	3206.37± 21.82	-0.47
RGUK-1	99.8% (potassium sulphate)	13600 - 14400	14000± 400	14089.47 ±83.82	0.22

The Gamma vision 32 software (manufacture by E&G Ortec company Tennessee 37831 USA) were used for analysis spectra. The concentration of radionuclides were determined in soil samples using the equation 2 [12]:

$$\text{Activity}(\text{Bq kg}^{-1}) = \frac{N-B}{T\times\rho\times\varepsilon\times\omega} \quad (2)$$

Where; T is the counting time (s), ρ is the gamma emission probability, ε is the absolute detector efficiency in the specified gamma energy and ω sample mass (kg), N number of the desired peak, and B is the background spectrum counting at the peak located at the same time. The average of specific activity and its uncertainty in all of soil samples were calculated by equations 3 and 4:

$$A(\text{average}) = \frac{\sum_{i=1}^{89}\frac{A_i}{\sigma_i}}{\sum_{i=1}^{89}\frac{1}{\sigma_i}} \quad (3)$$

$$\sigma(\text{average}) = \frac{1}{\sqrt{\sum_{i=1}^{89}\frac{1}{\sigma_i^2}}} \quad (4)$$

Where; A_i (Bq kg^{-1}) and σ_i are specific activity

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and its uncertainty, respectively [13]. The minimum detectable activity (MDA) was also determined from the background radiation spectrum using equation 5 [14]:

$$MDA(Bq\ kg^{-1}) = \frac{2.76+(4.6\times\sqrt{B})}{T\times\rho\times\varepsilon\times\omega} \quad (5)$$

The minimum detectable activity (MDA) for ^{137}Cs was found as 1.43 Bqkg^{-1}. The specific activity of the ^{226}Ra was determined using the gamma lines 295.2 keV and 351.9 keV for ^{214}Pb and 609.3, 1120.3 and 1764.3 for ^{214}Bi. For ^{232}Th, the specific activity was determined using the gamma lines 338.40 keV and 911.07 keV for ^{228}Ac. In the case of ^{40}K and ^{137}Cs, the specific activities were evaluated by their individual gamma lines of 1460.75 keV and 661.7 keV, respectively [12]. The worldwide average values of specific activity ^{226}Ra, ^{232}Th and ^{40}K are 30, 35 and 400 Bq kg^{-1} respectively[1].

Radiological parameters

Radium equivalent activities (Ra_{eq})

98.9% of the radiological effects of the uranium series come from ^{226}Ra and its daughters [12]. Therefore, to determine the total natural radioactivity of the soil and determine the level of environmental safety of radiation and its comparison with the maximum permitted content of radionuclides, the concept of radium equivalent is used. This concept is based on the fact that 10 Bq kg^{-1} of ^{226}Ra, 7 Bq kg^{-1} of ^{232}Th and 130 Bq kg^{-1} of ^{40}K gives the same dose of gamma radiation, respectively. The radium equivalent activity can be calculated using equation 6 [1]:

$$Ra_{eq} = A_{Ra} + 1.43\ A_{Th} + 0.077\ A_K \quad (6)$$

For building material this quantity should be less than 370 Bq kg^{-1} [15].

Absorbed dose rate (D)

Absorbed dose rate in air at height of one meter above ground (D) due to gamma rays emitted from ^{226}Ra, ^{232}Th and ^{40}K radioactive nuclei in soil is calculated using equation 7 [1]:

$$D(nGy\ h^{-1}) = 0.427\times A_{Ra} + 0.662_{Th} + 0.0432\times A_K \quad (7)$$

Dose rate absorption for the anthropogenic radionuclide ^{137}Cs, can be calculated using equation 8 [16]:

$$D^{\circ}_{Cs} = 0.03 \times A_{Cs} \quad (8)$$

Where; A_{Cs} is the specific activity of the ^{137}Cs and 0.03 is the dose conversion factor for the ^{137}Cs activity per Bq kg^{-1} so dose rate is expressed in the units of nGy h^{-1}.

Internal and external hazard Indices (H_{in}, H_{ex})

The internal hazard index refers to the carcinogen inhalation of radon gas, and to limit this risk, the maximum permitted radium activity would be halved. The external exposure to gamma rays in the study area is called external hazard index and equations 9 and 10 are used to calculate these indices, respectively [1]:

$$H_{in} = \frac{A_{Ra}}{185} + \frac{A_{Th}}{259} + \frac{A_K}{4810} \leq 1 \quad (9)$$

$$H_{ex} = \frac{A_{Ra}}{370} + \frac{A_{Th}}{259} + \frac{A_K}{4810} \leq 1 \quad (10)$$

It was established that the maximum Hex value should not exceed 1, which corresponds to the maximum permissible Ra_{eq} value for building materials equal to 370 Bqkg^{-1} [15].

Gamma index (I_γ)

I $_\gamma$ is used to evaluate the radiation of gamma hazard which associated with the natural radionuclide in studied soil samples. It was calculated by equation 11:

$$I_\gamma = \frac{A_{Ra}}{150} + \frac{A_{Th}}{100} + \frac{A_K}{1500} \quad (11)$$

Where; A_{Ra}, A_{Th}, A_K and A_{Cs} in equations 6 to 11 are specific activities of ^{226}Ra, ^{232}Th, ^{40}K and ^{137}Cs in Bqkg^{-1} [1]. It is a test tool for the identification of the material that may pose a health risk when it is used in the construction of buildings [17].

Annual effective dose equivalent (AEDE)

The effective dose equivalent to the absorption of gamma radiation throughout the

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year by an adult person depends on the time of exposure. According to a report prepared by UNSCEAR (2008), it is assumed that the urban population spends 80% of the time indoors, and spends twenty percent of their time outside. Therefore, the annual effective dose received by adult person can be estimated by using equation 12:

$$AEDE(mSv\ y^{-1}) = Q \times T \times W \times D \times 10^{-6} \quad (12)$$

Where; the AEDE represents annual effective dose equivalent in $mSvy^{-1}$, Q is the conversion factor as 0.7 $SvGy^{-1}$, T is the time in hours for one year, i.e. 8760 hours, D is the dose rate in $nGyh^{-1}$ and W is the occupancy factor for outdoor and indoor as 0.20 and 0.80 respectively [1].

Excess lifetime cancer risk (ELCR)

ELCR is an indicator that depends on the average annual effective dose received by adult person which is caused by gamma emissions from the ground. This parameter is calculated using equation 13:

$$ELCR = AEDE \times LT \times RF \quad (13)$$

Where; AEDE is annual effective dose rate, LT is average life of time with value 70 years and RF is risk factor in Sv^{-1}. For people, the International Commission on Radiological Protection (ICRP 60) established the values of RF as 0.05 Sv^{-1} [18].

RESULTS

The result of specific activities of natural and ^{137}Cs radionuclides measured for 39 samples included cultivated and virgin soil is presented in table 2. Also the results of the calculation of radiological parameters for cultivated and virgin soil samples presented in tables 3, 4 and 5.

Table 2. The specific activities of natural radionuclides and ^{137}Cs in $Bqkg^{-1}$. in cultivated and virgin soil samples.

Sample code	specific activities of radionuclides (Bq kg^{-1})				Sample code	specific activities of radionuclides (Bq kg^{-1})			
	^{226}Ra	^{232}Th	^{40}K	^{137}Cs		^{226}Ra	^{232}Th	^{40}K	^{137}Cs
Cultivated soil samples					Virgin soil samples				
CS1	19.98±1.52	23.44±1.80	403.51±10.15	5.14±0.43	VS1	22.16±1.80	27.73±1.47	523.19±17.64	8.66±0.95
CS2	20.76±1.84	29.46±2.70	354.12±11.48	6.91±0.64	VS2	17.83±1.26	22.11±1.34	367.36±9.57	<1.43
CS3	21.90±1.93	31.22±1.96	429.66±15.31	3.95±0.68	VS3	24.16±2.02	27.74±1.56	447.68±11.07	<1.43
CS4	24.55±1.44	26.58±1.5	415.03±10.41	4.10±0.42	VS4	23.86±2.13	25.51±1.39	415.27±10.38	<1.43
CS5	23.40±1.81	27.24±1.53	450.71±10.99	2.70±0.42	VS5	19.64±1.43	27.19±2.15	327.09±12.35	<1.43
CS6	12.53±1.63	14.32±1.3	241.06±7.516	2.14±0.39	VS6	22.87±1.83	31.92±1.82	483.17±11.56	5.68±0.69
CS7	20.93±1.87	27.25±1.58	433.16±10.74	4.52±0.59	VS7	23.00±1.81	30.70±1.35	494.03±11.69	1.84±0.39
CS8	26.98±1.75	37.87±1.40	502.65±14.82	5.26±0.76	VS8	26.19±1.70	36.09±2.36	520.42±12.23	5.30±0.43
CS9	14.82±1.32	19.65±1.19	309.87±13.92	5.20±0.93	VS9	26.94±2.09	37.67±1.70	529.96±12.45	3.00±0.42
CS10	22.00±1.66	30.94±1.40	566.01±17.16	1.04±0.38	VS10	25.56±1.35	45.75±1.74	569.13±18.39	4.96±0.79
CS11	13.46±1.30	19.52±1.30	344.34±12.34	<1.43	VS11	27.28±1.42	33.24±1.64	487.47±11.68	5.20±0.43
CS12	19.94±1.46	31.14±1.98	493.94±11.78	3.89±0.42	VS12	25.63±1.82	36.60±2.14	488.92±11.78	7.21±0.46
CS13	23.07±1.54	20.56±1.63	445.3±10.93	2.92±0.39	VS13	29.01±2.28	31.80±1.57	583.11±15.96	4.27±0.42
CS14	22.28±1.33	28.063±2.10	498.67±13.73	2.62±0.42	VS14	14.72±1.27	22.48±2.03	271.86±8.042	4.42±0.71
CS15	25.37±1.33	31.76±2.06	601.31±17.45	5.98±0.81	VS15	26.25±1.64	29.73±1.71	517.1±14.61	<1.43
CS16	25.61±2.40	26.93±1.4	442.09±10.81	3.21±0.40	VS16	30.15±1.90	36.66±1.59	589.39±15.35	3.47±0.43
CS17	19.84±2.34	36.77±2.22	519.64±12.2	4.26±0.41	VS17	30.05±1.45	34.96±1.58	597.27±17.46	17.78±0.82
CS18	18.61±1.83	28.85±1.33	548.11±17.15	<1.43	VS18	31.36±2.08	36.27±1.76	611.61±13.85	6.16±0.73
CS19	23.32±2.11	23.13±1.64	430.67±13.81	9.77±0.48	VS19	14.72±1.27	22.48±2.03	271.86±8.042	4.42±0.71
					VS20	29.61±1.83	50.32±3.21	565.02±18.23	10.12±0.92
mean	20.81±0.37	26.99±0.38	430.43±2.74	4.26±0.14	mean	23.99±0.37	31.74±0.38	461.09±2.68	5.51±0.14

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Table 3. Radiological parameters of soil samples.

Sample code	Ra_{eq}	H_{in}	H_{ex}	I_γ	Sample code	Ra_{eq}	H_{in}	H_{ex}	I_γ
	Cultivated soil samples					Virgin soil samples			
CS1	84.57	0.28	0.23	0.63	VS1	103.6	0.33	0.28	0.78
CS2	90.15	0.29	0.24	0.67	VS2	77.74	0.25	0.21	0.58
CS3	99.62	0.32	0.27	0.74	VS3	98.29	0.33	0.27	0.73
CS4	94.52	0.32	0.26	0.7	VS4	92.33	0.31	0.25	0.69
CS5	97.06	0.32	0.26	0.72	VS5	83.71	0.27	0.23	0.62
CS6	51.57	0.17	0.14	0.39	VS6	105.7	0.34	0.29	0.04
CS7	93.25	0.3	0.25	0.7	VS7	105	0.34	0.28	0.03
CS8	119.8	0.39	0.32	0.89	VS8	117.9	0.38	0.32	0.88
CS9	66.78	0.22	0.18	0.03	VS9	121.6	0.39	0.33	0.9
CS10	109.8	0.35	0.3	0.83	VS10	134.8	0.43	0.36	1
CS11	67.9	0.22	0.18	0.51	VS11	112.4	0.37	0.3	0.83
CS12	102.5	0.32	0.28	0.77	VS12	115.6	0.38	0.31	0.86
CS13	86.76	0.29	0.23	0.65	VS13	119.4	0.39	0.32	0.89
CS14	100.8	0.33	0.27	0.76	VS14	67.8	0.22	0.18	0.5
CS15	117.1	0.38	0.32	0.88	VS15	108.6	0.36	0.29	0.81
CS16	98.16	0.33	0.27	0.73	VS16	128	0.42	0.35	0.95
CS17	112.4	0.35	0.3	0.84	VS17	126	0.41	0.34	0.94
CS18	102.1	0.32	0.28	0.77	VS18	130.3	0.43	0.35	0.97
CS19	89.56	0.3	0.24	0.67	VS19	67.8	0.22	0.18	0.5
					VS20	145.1	0.46	0.39	1.07
mean	93.92	0.31	0.25	0.68	mean	108.08	0.35	0.29	0.73

Table 3. Radiological parameters of soil samples.

Sample code	Dose Rate ($nGyh^{-1}$)		$AEDE_{in}$ ($mSvy^{-1}$) For Cs $\times 10^{-3}$		$AEDE_{out}$ ($mSvy^{-1}$) For Cs$\times 10^{-4}$		ELCR$\times 10^{-3}$ For Cs $\times 10^{-6}$	
	$^{226}Ra,^{232}Th,^{40}K$	^{137}Cs	$^{226}Ra,^{232}Th,^{40}K$	^{137}Cs	$^{226}Ra,^{232}Th,^{40}K$	^{137}Cs	$^{226}Ra,^{232}Th,^{40}K$	^{137}Cs
CS1	40.21	0.15	0.20	0.76	0.05	1.89	0.18	0.66
CS2	42.15	0.21	0.21	0.1	0.05	2.54	0.18	0.89
CS3	46.89	0.12	0.23	0.58	0.06	1.45	0.21	0.51
CS4	44.70	0.12	0.22	0.60	0.05	1.51	0.18	0.53
CS5	46.06	0.08	0.23	0.40	0.06	0.99	0.21	0.35
CS6	24.49	0.06	0.12	0.31	0.03	0.79	0.11	0.28
CS7	44.19	0.14	0.22	0.66	0.05	1.66	0.18	0.58
CS8	56.30	0.16	0.28	0.77	0.07	1.90	0.25	0.67
CS9	31.64	0.03	0.16	0.14	0.04	0.34	0.14	0.12
CS10	52.46	N.D*	0.26	N.D*	0.06	N.D*	0.21	N.D*
CS11	32.37	N.D*	0.16	N.D*	0.04	N.D*	0.14	N.D*
CS12	48.62	0.12	0.24	0.57	0.06	1.43	0.21	0.5
CS13	41.65	0.09	0.20	0.43	0.05	1.08	0.18	0.38
CS14	48.04	0.08	0.23	0.38	0.06	0.96	0.21	0.34
CS15	55.98	0.18	0.27	0.88	0.07	0.22	0.25	0.08
CS16	46.53	0.1	0.23	0.48	0.06	1.20	0.21	0.42
CS17	53.04	0.13	0.26	0.62	0.07	1.57	0.25	0.55
CS18	48.88	N.D*	0.24	N.D*	0.06	N.D*	0.21	N.D*
CS19	42.70	0.29	0.21	1.44	0.05	3.60	0.18	1.26
mean	44.57	0.11	0.22	0.57	0.05	1.22	0.19	0.43

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Table 4. Other radiological parameters of cultivated soil samples.

Sample	Dose Rate ($nGyh^{-1}$)		$AEDE_{in}$ ($mSvy^{-1}$) For Cs $\times10^{-3}$		$AEDE_{out}$ ($mSvy^{-1}$) For Cs $\times10^{-3}$		ELCR$\times10^{-3}$ For Cs $\times10^{-6}$	
	$^{226}Ra, ^{232}Th, ^{40}K$	^{137}Cs	$^{226}Ra, ^{232}Th, ^{40}K$	^{137}Cs	$^{226}Ra, ^{232}Th, ^{40}K$	^{137}Cs	$^{226}Ra, ^{232}Th, ^{40}K$	^{137}Cs
VS1	49.46	0.26	0.24	0.13	0.06	0.32	0.21	1.11
VS2	36.92	N.D*	0.18	N.D*	0.05	N.D*	0.18	N.D*
VS3	46.58	N.D*	0.23	N.D*	0.06	N.D*	0.21	N.D*
VS4	43.75	N.D*	0.21	N.D*	0.05	N.D*	0.18	N.D*
VS5	39.14	N.D*	0.19	N.D*	0.05	N.D*	0.18	N.D*
VS6	50.00	0.02	0.25	0.01	0.06	0.03	0.21	0.09
VS7	49.77	0.01	0.24	0.01	0.06	0.01	0.21	0.05
VS8	55.60	0.16	0.27	0.08	0.07	0.19	0.25	0.68
VS9	57.30	0.09	0.28	0.04	0.07	0.11	0.25	0.39
VS10	63.17	0.15	0.31	0.07	0.08	0.18	0.28	0.64
VS11	53.01	0.16	0.26	0.08	0.07	0.19	0.25	0.67
VS12	54.34	0.22	0.27	0.11	0.07	0.26	0.25	0.93
VS13	56.93	0.13	0.28	0.06	0.07	0.16	0.25	0.55
VS14	31.72	0.13	0.16	0.06	0.04	0.16	0.14	0.57
VS15	51.65	N.D*	0.25	N.D*	0.06	N.D*	0.21	0.00
VS16	60.65	0.10	0.30	0.05	0.07	0.13	0.25	0.45
VS17	59.90	0.53	0.29	0.26	0.07	0.66	0.25	2.29
VS18	61.90	0.18	0.3	0.09	0.08	0.23	0.28	0.79
VS19	31.72	0.13	0.16	0.06	0.04	0.16	0.14	0.57
VS20	67.64	0.30	0.33	0.15	0.08	0.37	0.28	1.30
mean	51.06	0.13	0.25	0.06	0.06	0.17	0.22	0.55

*.Not Detected

DISCUSSION

Activity and statistical analysis of activity

The range of the specific activities of ^{226}Ra, ^{232}Th, ^{40}K and ^{137}Cs for cultivated (virgin) soil samples ranged from 12.53 ± 1.63 (14.72 ± 1.27) to33.03±1.79 (31.36 ± 2.08), 11.30±2.07 (22.11 ± 1.34) to 37.87±1.40 (50.32 ± 3.21), 271.86±8.04 (271.86±8.04) to 605.50±18.02 (611.61±13.85) and <1.43 to 13.36±0.89 (17.78±0.82) in $Bqkg^{-1}$, respectively. The average values of corresponding radionuclides calculated as 20.81±0.83 (23.99 ± 0.37), 26.99±0.38 (31.74 ± 0.38), 430.34±2.74 (461.09±2.68), 4.26±0.14 (5.51±0.14) in $Bqkg^{-1}$, respectively. The obtained results indicate that the average specific activity ($Bqkg^{-1}$) of ^{40}K and ^{232}Th in virgin soil samples was close to the global average, but in the case of ^{226}Ra it was lower [1].The statistical analysis of the data was performed using SPSS version 16. Using the Pearson linear regression method, the correlation coefficient R^2 for radium-potassium and potassium- thorium in cultivated soil (stellar) and virgin soil (square) are shown in figure 2. The amounts of R^2 (standard beta coefficient) were 0.555 and 0.864 for cultivated and virgin soils, indicating respectively a significant correlation between radium and potassium at a probability of 1%. This factor shows that cultivated soil irrigation will be caused by the radium salt transferred to the deep layer. Correlation coefficients between every two different radionuclides calculated by this method are listed in table 6. The strength of the correlation was described using a guide, which Evans (1996) suggested for the absolute value of R^2. The results showed a strong positive correlation between ^{226}Ra and ^{232}Th and very strong between ^{226}Ra and ^{40}K with values of 0.732 (0.693) and 0.934 (0.753), respectively in the virgin (cultivated) lands. In both methods, the values of the correlation coefficient obtained the same. There is an agreement between data

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in both of them. According to the data in table 6, the weak correlation between cesium and other radionuclides is evident because cesium as an artificial nuclide is transported by wind from other countries during nuclear accidents.

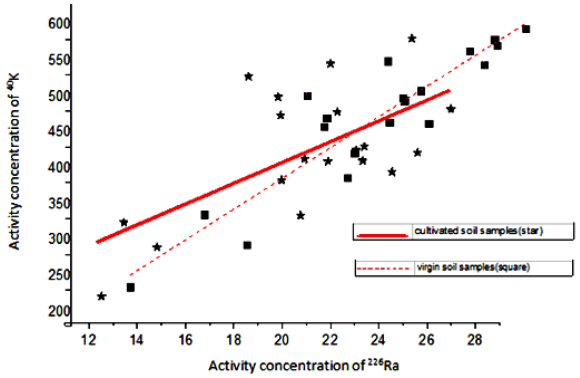


Figure 2. Variability of ^{40}K vs ^{226}Ra activity concentration in cultivated (star) and in virgin (square) soils.

Table 6. Correlation coefficients between ^{226}Ra and other radionuclides for cultivated and virgin soil.

radionuclide		linear statistical model			
		regression		Pearson	
independent variable	dependent variable	Standardized coefficients Beta	sig	Correlation coefficients	sig
Cultivated soil					
^{226}Ra	^{40}K	0.633	0.035	0.633**	0.004
	^{232}Th	0.619	0.013	0.619**	0.005
	^{137}Cs	0.370	0.000	0.370	0.119
Virgin soil					
^{226}Ra	^{40}K	0.934	0.097	0.934**	0.000
	^{232}Th	0.732	0.02	0.732**	0.000
	^{137}Cs	0.385	0.000	0.385	0.094

**Correlation is significant at the 0.01 level.

Radiological indices

The estimated Ra_{eq} values range is from 51.57 to 119.8 with an average of 93.92 in Bqkg^{-1} in the cultivated soil samples.

The average Ra_{eq} value for the virgin soil was 108.08 Bqkg^{-1} (range: 67.8-145.1). These values are lower than the permissible maximum value of 370 Bqkg^{-1} and worldwide average (139.70). The calculated dose absorption in air at a height of 1 meter above the ground for samples is from 24.49 to 56.30, with average values of 44.57 and 51.06 respectively in nGyh^{-1}. In samples of cultivated and virgin soil, these values were lower than worldwide average as 55 nGyh^{-1} [1].The mean value of absorbed dose in air due to ^{137}Cs was calculated, respectively, 0.11 and 0.13 in nGyh^{-1}. The AEDE indoor absorbed by an adult due to natural and artificial radionuclides ranged from 0.12 to 0.28, and ND to 1.44 × 10^{-3}, on average 0.22 and 0.57 × 10^{-3} in mSvy^{-1} for cultivated soils, while the average of this index for outdoor was 0.05 and 1.22 × 10^{-4} mSvy^{-1}, respectively. The ranges of AEDE indoor due to natural and artificial radionuclides in virgin soil were calculated as 0.16-0.33 and ND-2.61× 10^{-3} in mSvy^{-1} with average values of 0.25 and 0.63 × 10^{-3} mSvy^{-1}, respectively, while the average of this index for outdoor is 0.06 and 1.67 × 10^{-4} mSvy^{-1}. The average global value is 0.48 mSvy^{-1}, of which 0.07 mSvy^{-1} comes from outside and 0.41mSvy^{-1} from indoor [1]. Therefore, the area does not pose a threat to the population. The average annual dose of ^{137}Cs gamma radiation for all samples is less than the reported 0.6 mSvy^{-1} values by the United Nations Scientific Committee on the effects of atomic radiation [1]. The external hazard index deals with the assessment of excess gamma radiation from radionuclides in cultivated or virgin soils. Average H_{in} and H_{ex} values for cultivated (virgin) soils samples obtained as 0.31 (0.35) and 0.25 (0.29) respectively, which are lower than the maximum allowable limit [1]. In the case of virgin soil, the indicators of external and internal threats are higher than that of cultivated land, which shows that radium and thorium salts can be transported to deeper layers by irrigation. The index of I_γ is ranged from 0.39 to 0.89 and from 0.03 to 1.07, with an average value of 0.68 and 0.73 for the cultivated and virgin soil samples, which for most of them are less than one value. Table 5 lists the specific activities of natural radionuclides in cultivated and primary soils of some countries, which shows that in Pakistan, Turkey, Bangladesh and Malaysia are higher and for other countries are in the same range. Distribution maps of ^{226}Ra, ^{232}Th and ^{137}Cs in Fig. 3 for cultivated land were plotted using the SURFUR software version 15, which shows that the ^{226}Ra and ^{137}Cs distributions are similar. Since the distribution of ^{137}Cs depends on the wind and the

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topography of the area, these maps show that part of the radium is dispersed in environments by fly ash from the outlet of the chimneys of refinery by the wind. The excess lifetime cancer risk (ELCR) was obtained for cultivated (virgin) soil as 0.19 × 10^{-3} (0.22 × 10^{-3}), which is in the range to the world average (0.29 × 10^{-3}) and is less than the permissible value (10^{-3}). Table 7 provides a comparison of the average activity of natural radionuclides in Bqkg^{-1} for some countries, which show that in Pakistan [16], Turkey [19, 24], Bangladesh [22], and Malaysia [23], are higher and for other countries are in the same range.

Table 7. Comparison of the average activity of natural radionuclides in Bqkg-1 for some countries.

country	^{238}Ra	^{232}Th	^{40}K	reference
cultivated soil samples				
Syria	23	20	270	(1)
Oman	29.7	15.9	225	(2)
Pakistan	73.9	152.2	325.3	(16)
Turkey	85.75	51.08	771.57	(19)
Iran(Arak and Sareband)	45.54	69.09	926.71	(20)
Iran	20.81	26.99	430.43	This work
virgin soil samples				
Bangladesh	60.2	60.8	928	(21)
china	38	57.6	838	(22)
Malaysia	51.06	78.44	125.66	(23)
Turkey	115	192	1207	(24)
Nigeria	18	22	210	(25)
Botswana	34.8	41.8	432.7	(26)
Yemen	44	58	822	(27)
Iran(Arak and Sareband)	37.27	43.18	604.05	(20)
Iran	23.99	31.74	461.09	This work

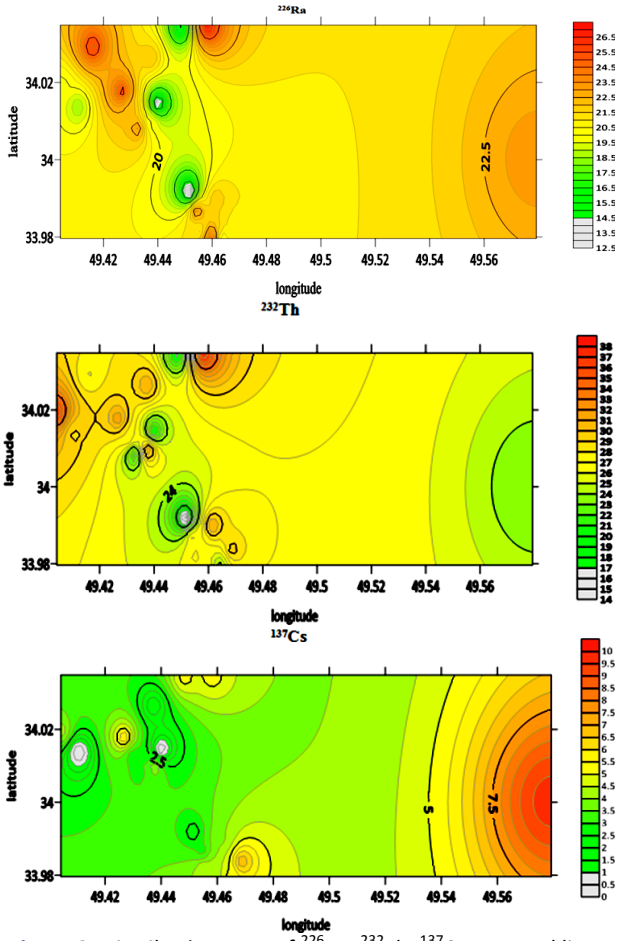


Figure 3. Distribution map of ^{226}Ra, ^{232}Th, ^{137}Cs on equal lines in cultivated studied regions (X-axis; longitude (E-W (Deg)), Y-axis: latitude(S-N (Deg))).

CONCLUSION

- Average activity concentration of radionuclides ^{226}Ra, ^{232}Th, ^{40} K and ^{137}Cs were measured which lies within the world range values in cultivated (virgin) soil samples.
- All soil samples were polluted by ^{137}Cs which indicate the pollution this region by radioactive dust originated from other countries.
- The average activity concentrations of the measured radionuclides in the virgin soils were higher than the cultivated soils, which may be due to the irrigation of the area during cultivation and the transfer of their salts to deeper layers.
- All calculated radiological parameters are within acceptable limits and do not threaten the dangers of the population living in this area.

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***Conflicts of interest*: Declared none.**

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